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Large Eddy Simulations and Direct Numerical Simulations of High Speed Turbulent Reacting Flows

by

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Abstract

The basic objective of this research is to extend the capabilities of *Large Eddy Simulations* (LES) and *Direct Numerical Simulations* (DNS) for the computational analyses of high speed reacting flows. In the efforts related to LES, we have been primarily involved with assessing the performance of various modern methods based on the Probability Density Function (PDF) methods for providing closures for treating the subgrid fluctuation correlations of *scalar* quantities in reacting turbulent flows. In the work on DNS, we have been concentrating on understanding some of the relevant physics of compressible reacting flows by means of statistical analysis of the data generated by DNS of such flows. In the research conducted in the second year of this program, our efforts have been focused on the modeling of homogeneous compressible turbulent flows by PDF methods, and on DNS of non-equilibrium reacting high speed mixing layers. Some preliminary work is also in progress on PDF modeling of shear flows, and also on LES of such flows.

1 Introduction

The need for the use of advanced computational methods for the analysis of high speed reacting flows is obvious [1, 2, 3]. Within the past thirty years, NASA Langley Research Center has been at the forefront in making use of advanced computational methods for the investigations of high speed turbulence, with and without the complex effects of chemical reactions. An overview of the work conducted at the Theoretical Flow Physics Branch reveals the extent of progress made by the scientists at NASA LaRC in this area of research.

One of the recent contributions by NASA in the field of high speed combustion is due to Drummond [4, 5, 6]. This work, consisting of the first systematic study of high speed reacting shear flows, paved the way for the subsequent contributions made in this research area within the past few years. Since this early work, there have been extensive contributions made by other investigators on various aspects of high speed combustion. The extent of these achievements and the need for even further extensions have been so great that recently NASA in collaboration with ICASE, organized a workshop focused on identifying various means by which the area of high speed combustion can benefit from the current state of knowledge in the "simpler" field of low speed combustion. A proceeding of this workshop is available [7]. As asserted at the conclusion of this workshop, one of the major issues of crucial interest to NASA-LARC is the need for further developments and more substantiated work in utilizing advanced computational methods for the analysis of compressible turbulent combustion.

A review of immediate needs in computational treatment of high speed flows is available [1]. According to this review, there is a need for further utilizations and improvements of the methods known as Direct Numerical Simulations (DNS) and Large Eddy Simulations (LES) for the computational treatment of compressible reacting turbulence. These two types of simulations have been labeled by this PI as "Model Free Simulations" [8], to emphasize the distinction between these methods and those based exclusively on "turbulence models" [9, 10]. Model free simulations have been the subject of wide utilizations by various groups

at NASA LaRC. This consists of contributions by both DNS and LES. Since the work of Drummond [4, 5], and the development of the computer code *SPARK*, the extent of achievement has been promising. Some of the most recent contributions in this area are those in [11, 12, 13]. The SPARK code has been the subject of validations and improvements by various investigators at TFPB, NASA LaRC. Currently with the developments of modern numerical algorithms [14], advanced turbulence closures [15, 16], modern grid generations procedures [17], this code is being utilized as an efficient tool with accurate predictive capabilities for a variety of practical flow systems. This is clearly evident by the enthusiasm of the Aerospace industry in making use of the capabilities facilitated by this code [18].

There has also been a substantial interest at TFPB, NASA LaRC in developing and implementing LES for the treatment of compressible turbulent flows. This goers back to the early work of Erlebacher *et al.* [19], and since then LES have been the subject of wide utilizations by many scientist at this branch and also at ICASE [20, 21, 22]. Needless to indicate that the "Transition Group" at this branch is credited as the leader within the international scientific community in LES of compressible and transitional flows [23].

The outcome of earlier investigations and also those in progress clearly suggest that foreseeable developments of advanced computational facilities will not be sufficient to relax the restriction of DNS to flows having small to moderate variations of the characteristic length and time scales. This is not an unknown problem, and has been well-documented in turbulence literature (for recent discussions see [24, 25, 8, 26]). The notorious limitations associated with the direct simulation of high Reynolds number flows clearly indicates that the boundaries of applicability of DNS are, and will continue to be, somewhat restricted. Nevertheless, within its domain, DNS can be used to enhance our understanding of the physics of turbulent flows by (1) providing specific information concerning the detailed structures of the flow, and (2) providing a quantitative basis for evaluating the performance of turbulence closures.

LES appear to provide a good alternative to DNS for computing flows having ranges of

parameters similar to those encountered in practical systems [27, 28, 29, 30, 31, 32, 33, 22, 25, 8, 26]. The approach based on LES has a particular advantage over the turbulence modeling procedures in that only the effects of small-scale turbulence motion have to be modelled. Therefore, the construction of accurate "subgrid scale" closures is an important task in its implementation. The extensive experience gained during the past two decades in constructing turbulence models for the Reynolds-averaged equations of turbulent combustion have proven useful in the development of these closures. A major advantage offered by LES is that, subgrid closure modeling can be substantially simplified by performing computations over grids of different size [19, 21]. In this setting, the performance of a model on coarse grids can be directly evaluated by comparing its predictions with those obtained on fine grids. This procedure has been followed in many previous works [28, 19, 21, 20, 34] and the simulations, in many cases, have produced satisfactory subgrid closures (See [23] for a review of recent achievements). Most of these efforts have been towards constructing closures for non-reacting flows [30, 31], and it has been only recently that, the LES has been introduced for the analysis of reactive phenomena [34].

In the work conducted under this grant, we have continued our investigations on both LES and DNS related to compressible reacting flows. In the next section, the factors motivating research in the format proposed here are given, and in Section 3 a summary is provided of our accomplishments with this support at the conclusion of the second year of the research. The publications resulting from our efforts in Year 2 are listed in Section 3, together with the Awards and Honors received by the members of the research team supported by this Grant.

2 Motivation

The physics of high speed reactive flows is rich with many complexities [35], some of which the subject of intense investigations since the Apollo Project. A few examples of the physical

issues of current interest are the questions associated with the chemical and thermodynamics non-equilibrium effects, the cause and effect of turbulence, the interactions of turbulence and chemistry, the real gas effects at high temperatures, *etc.* This complexity necessitates investigations in many diverse areas consisting of all the branches of engineering. Having the discussions limited to technical issues, one of the most important elements of current interest is associated with the phenomenon of high speed turbulent combustion in internal flows [1]. Even in this relatively limited range, the physical complexities associated with "turbulence" is so broad that requires collaborative interactions amongst scientists within different disciplines in dealing with its cause and effects.

An important tool in the analysis of high speed flows, as recognized by today's Aerospace research community, is Computational Fluid Dynamics (CFD) [36, 37] (also see [38]). The power and the extent of capabilities of modern supercomputers have been instrumental in our efforts towards addressing and solving some of the existing problems in this field.

As a result of collaborative efforts between SUNY-Buffalo and NASA-LaRC, we have been able to identify, and work on some important aspects of CFD utilizations in high speed combustion research. The particular aspect of the problem under investigation, in accord with our expertise, involves model free simulations of high speed turbulent combustion. Having the general guidelines and directions set by NASA-LaRC scientists, we initiated this work aimed at understanding several important issues in this direction. Our specific objectives from the start of this research has been to: (1) develop reliable mathematical closures for LES of high speed reacting flows, and (2) take advantage of the capabilities of DNS for further understanding of the physical issue associated with high speed transport. Our primary focus has been on the first task, while necessarily some efforts had to be devoted to the second task.

In the next section, the summary of our to-date achievements are described. A more detailed descriptions of our achievements can be found in Appendix I through Appendix III.

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3 Summary of Our Accomplishments to Date

The subject matters discussed in this section are focused on both of the basic elements of the proposed research. Therefore, here they are discussed separately in order.

3.1 LES and DNS of Homogeneous Reacting Turbulence

It is now widely suggested that in developing a reliable subgrid closure for LES of turbulence, it is useful to consider a flow at the simplest possible condition. Namely, a homogeneous flow, with or without a spatial isotropy. This type of flow has been considered in almost all the previous works towards the developments of subgrid closures for LES [39, 40, 41, 42, 19, 21, 20, 43]. The factors justifying this choice are based on the simple fact that the flow within the subgrid can be assumed isotropic regardless of the nature of the large scale behavior. Therefore, with the development of a turbulence model in a homogeneous setting, the generated results can be directly used in flows with strong spatial non-homogeneities. This line of reasoning is justified if the filtered portion of the transport variable constitute a sufficiently large portion of the "whole" flow, so that the assumption of spatial homogeneity within the un-filtered part is valid.

In almost all previous work in developing subgrid closure for non-reacting flows, the approach based on "Moment Methods" has been adopted. The essential element of such methods is based on the revisions of the well-known Smagorinnsky [44] model (also see [45, 46]) which is somewhat analogous to the widely used gradient diffusion type closures in turbulence modeling [9]. The major modifications include the possible means by which the physical effects, such as compressibility, *etc.* are incorporated into the model. The extent of progress made to-date indicates that the state of knowledge necessary to incorporate such effects is not yet at a maturity level to justify absolute validity. For example, as indicated in [20], currently, only flows with low to moderate ranges of compressibility can be considered, and

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the methodology is not capable of simulating flows at very high compressibility levels.

Unfortunately, moment methods have not proven very effective in the modeling of turbulent reactive phenomena [47]. The results of extensive work within 1970's-1980's clearly reveals the shortcomings of such models primarily because of the lack of acceptable closures for all the ranges of the Damköhler number. Depending on the nature of the closure, a model which is reasonable in a "frozen flow" cannot be used for modeling of a chemically reacting flow under "equilibrium conditions," and vise versa. This problem stems due to the drawbacks of the model to account for the effects of higher order unclosed moments at a selected level of truncation.

The problem discussed above can be circumvented, to a large extent, by following the approach based on the Probability Density Function (PDF) methods [48, 49, 50, 51, 8, 52]. The advantage of PDF methods in the modeling of reactive flows is due to the simple fact that with such models, all the contributions of the higher order moments are recovered by a single PDF transport equation. Therefore, once a model is developed there are no needs for constructing additional closures. The pleasing feature is that, this model can be developed in the simpler case of a non-reactive flow. Since the effects of chemical reactions appear in a closed form, the generated model can be subsequently used for the modeling of reactive phenomena [49, 51, 52, 53, 50, 8].

There are two general ways by which the PDF methods can be employed for solving turbulent combustion problems [10, 54, 8]: (1) Assuming the form of the PDF [48], (2) Solving a transport equation for the PDF [55, 56, 57]. Both of these approaches are well-known in combustion literature [15] and will not be elaborated upon here in detail. In the first approach, the PDF is parameterized with the knowledge of its first few (usually the first two) moments. This approach is obviously easier, but the moments must be provided by turbulence models. In the second approach, the consideration of the PDF transport equation is required. The problem with this approach, in addition to being more computationally demanding, is that some modeling is required to account for the the effects of the PDF

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transport in the domain of the scalar composition.

Knowing the general state of progress in PDF methods in turbulence modeling, we decided to examine both these PDF approaches in LES of turbulent reacting flows. That is, we initiated a parallel research program in which the potential capabilities and the limitations of both these methods were examined. In doing so, in many cases, we had to first focus on some of the properties of these models before being able to use them for the purpose of developing subgrid closures. Here, a summary is provided of all our findings in this aspect of our work:

1. In the context of single-point PDF formulation, currently the Amplitude Mapping Closure (AMC) of Kraichnan [58, 59, 60, 61, 62, 63, 64] provides the most satisfactory results in the approach based on the PDF transport equations. This approach has proven more satisfactory than many of the previous models based on the so-called Coalescence/Dispersion (C/D) closure [65, 66, 50, 57, 67] in providing an acceptable evolution for the PDF transport.

2. The main deficiency of the AMC (or any other single-point PDF model) is associated with its incapability in predicting the frequency (or length) scale of PDF evolution. The EDQNM (Eddy-Damped Quasi Normal Markovian) spectral closure [68] provides a reasonable means of overcoming this deficiency. In short, EDQNM provides a transport equation for the "covariance" of the scalar variable $\rho(\underline{r})$ as a function of the "length scale." The advantage of this closure is its capability of including the length scale information (through the parameter r) into the formulation. From this viewpoint, the closure is much more powerful tan the typical $K - \epsilon$ [9] type closures. However, presently the use of the closure is limited to homogeneous and incompressible flows.

3. The AMC is very effective in modeling of the mixing problem in homogeneous flows. Therefore, it is strongly recommended for statistical treatment of homogeneous reacting flows under chemically frozen and chemical equilibrium conditions. The use of the model is very convenient for the treatment of initially segregated reacting systems. In stoichiometric

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mixtures, the limiting rate of fuel consumption in a chemical reaction of the type *Fuel +*
Air → *Products*, the solution generated by AMC is of a simple algebraic form, and is very
convenient for practical applications [48, 69, 70, 71]. However, it is not very useful for LES,
because the reacting flow within the subgrid can hardly ever be assumed stoichiometric,
albeit homogeneous [34]. The extension of the model to account for non-unity equivalence
ratios has been made, but the final solution can be only expressed in terms of the integrals of
"Degenerate Hypergeometric Functions." In both cases, the predicted results compare very
well with DNS results and are better than any other closure developed in the literature within
the past forty years. Please see Appendix I for a detailed discussion of our achievements in
this aspect of our work .

4. The approach based on AMC requires a transport equation for the PDF. Therefore, its
applications for LES requires numerical solution of an additional transport equation. It is
shown that, in some circumstances, the family of probability frequencies of *Pearson Type*
[72] provides a simple alternative to AMC. Namely, the β-density of the first kind performs
reasonably well in approximating the single point PDF of a conserved scalar property, if the
evolution of its variance is known *a priori.* With the implementation of this density, it is
also possible to develop a closed form algebraic solution for the rate of reactant conversion as
indicated above. For a stoichiometric mixture the results are in terms of Gamma functions.
For non-stoichiometric mixtures, the results can only be expressed in the form of Incomplete
Beta Functions [73]. A mathematical justification has been provided of the similarity of the
Pearson PDF's with AMC generated frequencies. This justification fails if the PDF is not of
a bimodal type. To the best of our knowledge, the extension of Pearson type distributions
for the analysis of more complex distributions, is the subject of current research within the
communities of statisticians and biometricians (see Appendix I).

5. The use of the β density model has been extended for the modeling of non-homogeneous
reacting flows. The simulated results are very encouraging provided that the evolution of
the first two moments of the PDF are furnished externally. Again, the same limitations

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indicated above, holds. Appendix II provides a complete description of our achievements in this aspect of our work.

6. The Pearson family have been used for *a priori* analysis of subgrid modeling of the reacting flows in the limit of an infinitely fast chemical reaction. Our results suggest that in the absence of a better alternative, the *beta* density performs reasonably well in depicting the small scale behavior. However, as before, this does not advocate the generality of this density for practical applications. The results form this part of our work has been reported in [34], and has been included in our previous annual report to NASA LaRc (May 1991).

3.2 DNS of High Speed Reacting Mixing Layers

In the wake of our earlier works [12, 74, 13], we have continued our investigation of several issues of pertinence to the analysis of compressible reacting turbulence. This study has been motivated, to a large extent, in providing a computational complement to laboratory investigations of turbulent diffusion flames [75, 76]. We feel that, this is very important in view of the significant progress made within the past decade in the development and implementation of DNS for the analysis of unpremixed reacting flows [77, 78, 79, 80, 81, 82, 83, 84, 85, 12] (see [86, 87, 1, 88, 89, 3] for recent reviews). The extent of progress made by these contributions has been very encouraging, thus justifying further utilization of the direct methods in the analysis of turbulent flames.

As indicated before, in this aspect of our work, we were able to make use of present computational capabilities provided by the SPARK code. In this period, we also became involved in utilizing other computer codes for the sole reason of making comparative assessments of the SPARK code, as well as enhancing our knowledge of the state-of-the-art in this subject. Based on our work in the second year, we have been able to make the following solid conclusions:

1. With an idealized chemistry model of the type $Fuel + Air \rightarrow Product + Heat$, many fundamental issues in regard to the compositional structure of unpremixed spatially developing reacting mixing layer can be investigated. More specifically, the results of our simulations do portray the same qualitative behavior as those of recent laboratory measurements [90, 91, 92, 93]. Appendix III provides a complete description of our achievements in this aspect of our work.

2. For the first time, we have shown that exothermicity may actually act as to *increase* the rate of product formation in compressible reacting flows. This observation is not consistent with any of the previous DNS [94, 12] or experimental [95] results. The reason for this discrepancy is due to the effects of the Arrhenius kinetics model in our forced simulations and *cannot* be observed in the constant rate kinetics simulations and laboratory experiments [94, 12, 95]. This new observation, therefore, suggests that in the setting of a "turbulent" flame in which the "transitional effects" are not dominant, and where the chemical reaction is describable by an Arrhenius model, the heat release results in a *higher* reactant conversion, even though the rate of mixing may be somewhat less. Based on our observation, we recommend that the effects of exothermicity be assessed by means of laboratory measurements. These measurements must involve a reacting system whereby the rate of reaction conversion is temperature dependent (unlike that employed in [95]), and in which the large scale mixing intensity is not significantly affected by the heat release (see [94, 83, 88, 89, 84, 3] for discussions on the effects of heat release in low speed combustion, and [12, 74, 13] for discussions of such effects in high speed combustion). It is important to note that the issue would not be resolved by a mere comparison of the mixing characteristics between a reacting and a non-reacting layer. An appropriate analysis requires the consideration of two reacting systems with the same hydrodynamic characteristics but with different magnitudes of the enthalpy of combustion. The extent of validity of our conclusion under more complex chemistry models can be determined by these experiments (see Appendix III).

3. The result of our preliminary work in progress indicates that the concept of *Laminar*

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Diffusion Flamelets [96, 97] does reasonably well in the modeling of low speed unpremixed reacting flows. That is the results generated by DNS of a turbulent non-equilibrium flame, *scales* well with the *analytic* solution of an opposed laminar jet configuration in relating the generated products to the instantaneous dissipations of the scalar field. However, it is concluded that in moderate chemical reactions, the "scatter" of the DNS results is substantially more than that to be completely described by the flamelet model. Please see Appendix III.

4   Publications and Honors

Within the second year of this research, two journal publications and one conference papers have resulted form this work. The publications are identified in the Reference list by numbers [98, 73]. The conference paper is identified by number [99].

Within the past year, one of the graduate students involved in this research, Mr. Steven Frankel, was named as the second place winner in the "Technical Paper Presentation" of the *AIAA Northeastern Regional Conference*. We would like to mention that the first winner in this part of the completion was Mr. Richard S. Miller, one of the other graduate students currently in our research group at SUNY-Buffalo. Also, in the "Technical Presentation" (no paper) part of this conference, Ms. Francine Battaglia (who also work with P. Givi), was named as the first prize winner.

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5 Appendix I

In this Appendix, we discuss the properties of the Amplitude Mapping Closure the Pearson Family of distributions for predicting the limiting rate of reactant conversion in homogeneous turbulent flows. The results provided in this appendix are useful in several regards:

1. For the first time in forty year, closed form algebraic relations are obtained for predicting the maximum rate of reactant conversion in homogeneous turbulent flows. The major attraction of these relations is due to their applicability in both stoichiometric and non-stoichiometric (fuel-lean and/or fuel-rich) mixtures. As shown in this appendix, the results simplify substantially for unity equivalence ratio. The superiority of our results can be appreciated by a review of the most recent attempts in the modeling of homogeneous reacting turbulent flows (In Ref. [101], a review is presented of all the other closures available before this work). purpose).

2. A direct consequence of our mathematical results is their applications for subgrid scale modeling in equilibrium reacting flows. As we have indicated in our previous Annual Reports to NASA LaRC, the flow within the subgrid is hardly ever stoichiometric. That is, the equivalence ratio within the subgrid is strongly dependent on the large scale mixing behavior. Therefore, it is very pleasing to have a closed form relation to predict the limiting rate of reactant conversion at all equivalence ratios.

3. There is a careful discussion in this Appendix in extending this work for the modeling of non-equilibrium reacting flows with the implementation of multivariate statistical analyses.

The extension of this work for predicting non-homogeneous flows are presented in Appendix II.

The major part of this Appendix was prepared by Dr. Cyrus K. Madnia.

Reactant Conversion in Homogeneous Turbulence: Mathematical Modeling, Computational Validations and Practical Applications

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Abstract

Closed form analytical expressions are obtained for predicting the limiting rate of reactant conversion in a binary reaction of the type $F + rO \rightarrow (1 + r)$ *Product* in unpremixed homogeneous turbulence. These relations are obtained by means of a single point Probability Density Function (PDF) method based on the *Amplitude Mapping Closure* (Kraichnan, 1989; Chen et al., 1989; Pope, 1991). It is demonstrated that with this model, the maximum rate of the reactants' decay can be conveniently expressed in terms of definite integrals of the Parabolic Cylinder Functions. For the cases with complete initial segregation, it is shown that the results agree very closely with those predicted by employing a Beta density of the first kind for an appropriately defined Shvab-Zeldovich scalar variable. With this assumption, the final results can also be expressed in terms of closed form analytical expressions which are based on the Incomplete Beta Functions. With both models, the dependence of the results on the stoichiometric coefficient and the equivalence ratio can be expressed in an explicit manner. For a stoichiometric mixture, the analytical results simplify significantly. In the mapping closure, these results are expressed in terms of simple trigonometric functions. For the Beta density model, they are in the form of Gamma Functions. In all the cases considered, the results are shown to agree well with data generated by Direct Numerical Simulations (DNS). Due to the simplicity of these expressions and because of nice mathematical features of the Parabolic Cylinder and the Incomplete Beta Functions, these models are recommended for estimating the limiting rate of reactant conversion in homogeneous reacting flows. These results also provide useful insights in assessing the extent of validity of turbulence closures in the modeling of unpremixed reacting flows. Some discussions are provided on the extension of the model for treating more complicated reacting systems including realistic kinetics schemes and multi-scalar mixing with finite rate chemical reactions in more complex configurations.

1

1 Introduction

For the past forty years, since the early work of Hawthorne *et al.* (1949), estimation of the reactant conversion rate has been the subject of wide investigations in mathematical modeling of turbulent reacting flows. In unpremixed reacting systems, including diffusion flames, there are two factors by which this rate is influenced: (1) the speed at which the reactants are brought into the reaction zone, and (2) the rate at which they are converted to products through chemical reactions. These two mechanism are coupled through the mutual interactions of the fluid dynamics and the chemistry. The relative importance of the two mechanisms is characterized by the ratio of their appropriate time scales. This ratio is known as the Damköhler number (Da), and quantifies the characteristic frequency of the chemical reaction to that of the hydrodynamics.

The role of the Damköhler number in the characterization of reacting flows is very important (Williams, 1985). As the magnitude of this number increases, the influence of chemical reactions become more pronounced and the reactants decay at a faster rate. This rate is bounded at the upper limit as the Damköhler number becomes infinitely large ($Da \rightarrow \infty$). In this limit, the rate of reactant consumption is governed by the hydrodynamics, *i.e.* the reaction is "mixing controlled" and is determined by the speed at which the reactants are brought into an infinitely thin reaction zone (Bilger, 1980).

Obviously, with the assumption of an infinitely fast chemistry, it is not possible to account for many interesting issues associated with non-equilibrium effects in unpremixed flames (Libby and Williams, 1980). However, as indicated in the original pioneering work of Toor (1962), and later by O'Brien (1971); Bilger (1980), it is very important to have a prior estimate of this limiting rate in practical modeling of reacting flow systems. This is simply due to the fact that in this limit, the problem reduces to the simpler problem of "mixing" in which its analysis is far less complicated than that of an equivalent reacting system. Furthermore, it provides an accurate estimate for those reacting systems in which the chemical reaction time is much smaller than the characteristic time associated with the hydrodynamics.

In most analysis of turbulent reacting flows, the "statistical mean" value of the reactant conversion rate is of practical importance, and development of an appropriate turbulence model which can predict this mean rate has been the subject of extensive investigations (for reviews see Toor (1975); Brodkey (1981); Libby and Williams (1980); Williams (1985)). Amongst the theoretical tools developed, it is now firmly accepted that the approach based on the single-point Probability Density Function (PDF) of the scalar quantities is particularly useful, and this approach has been very popular in modeling of the reactant conversion in a variety of turbulent reacting flow systems (Pope, 1991; Kollmann, 1990; Pope, 1985; Pope, 1990). The advantage of PDF methods is due to its inherent capability to include information for all the statistical correlations amongst the scalar field. Therefore, once the PDF (or the joint PDF) of the scalar variables is determined, all the relevant statistics of the field are available without a need for additional closures.

The most logical and systematic means of determining the PDF involves the solution of an appropriate transport equation governing its evolution in a given reacting flow. In this equation, due to the nature of the formulation, the effects of chemical reaction appear in a closed form (Pope, 1976), regardless of its degree of complexity. However, the influences of molecular action cannot be fully described, and can be treated only by means of employing an appropriate closure. As noted by Pope (1991), in most previous applications, this problem has been circumvented through the use of the Coalescence/Dispersion (C/D) models. Examples of such models are the early C/D prototype of Curl (1963), the Linear Mean Square Estimation (LMSE) theory (O'Brien, 1980), the closure of Janicka et al. (1979), among others (Pope, 1982; Pope, 1985; Kosaly and Givi, 1987; Givi, 1989). Despite their advantageous characteristics, the shortcomings associated with the C/D closures in the probabilistic description of scalar transport are well recognized. Namely, none of the aforementioned models predict an asymptotic Gaussian distribution for the PDF of a conserved scalar variable in homogeneous turbulence (Pope, 1982; Kosaly and Givi, 1987; Pope, 1991); and those which are capable of doing so (e.g. Pope (1982)), do not predict the initial stages of mixing correctly (Kosaly, 1986).

Recent development of the Amplitude Mapping Closure by Kraichnan and co-workers (Kraich-

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nan, 1989; Chen *et al.*, 1989) (see also Pope (1991)) has provided a promising way of alleviating some of the problems associated with the C/D closures. This closure, in essence, provides a means of accounting for the transport of the PDF in composition space, and its validity and physical applicability have been evidenced in a number of comparisons against data generated by means of both direct numerical simulations (Pope, 1990; Pope, 1991; Gao, 1991a; Gao, 1991b; Madnia *et al.*, 1991b; Jiang *et al.*, 1992), and laboratory experiments (Frankel *et al.*, 1992). These results suggest that, at least in the setting of an isotropic turbulent flow, this closure has some superior features over all the previous C/D type models.

Based on this demonstrated superiority, our objective here is to further examine the properties of this closure and to assess its capabilities for applications in modeling of unpremixed turbulent reacting flow systems. In particular, it is intended to provide a reasonably simple recipe that can be used in conjunction with this closure for predicting the limiting rate of mean reactant conversion. However, since this is the first study of this type, and due to mathematical complexities (that soon become apparent), we have made some simplifying assumptions, which are indicated here at the outset. Firstly, we consider an idealized irreversible binary reaction of the type $F + rO \rightarrow (1 + r)$ *Product* with initially segregated reactants (F and O). In accordance with the discussions above, only the maximum rate of reactant conversion is considered, implying that the magnitude of the Damköher number is infinitely large. Secondly, the turbulence field is assumed statistically homogeneous. Thirdly, all the chemical species are assumed to have identical and constant thermodynamic properties. Finally, the flow field is assumed isothermal in which the dynamic role of the chemical reaction on the hydrodynamic field is ignored.

With all these assumptions, the reacting system considered is obviously an idealized prototype of conventional combustion systems. However, it does provide a good model for dilute reacting systems in typical mixing controlled plug flow reactors (Toor, 1962; Bilger, 1980; Toor, 1975; Hill, 1976; Brodkey, 1981). Moreover, because of the mathematical complexities, even in this simple case, it is deemed necessary to analyze this simplified system before considering more complex scenarios. Nevertheless, the model is capable of accounting for arbitrary values of the stoichiometric coefficients and for any equivalence ratio. This allows

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capturing of many interesting features, as will be demonstrated. Some issues regarding future extension of this work will be discussed. At this point, it is sufficient to indicate that the approach followed here can be directly extended for treating more complicated flows with relaxation of all of the assumptions made here. However, in those cases, the final results cannot be generally expressed in terms of simple mathematical relations, and require numerical solution of the governing equations. Pope (1991) describes the procedure by which the transport equations generated by the mapping closure can be treated numerically.

For the idealized case of initially segregated reactants F and O, as indicated above, the initial marginal PDF's of their mass fractions are composed of "delta functions". Therefore, it is also speculated that the approach based on an *assumed* probability distribution may also provide a reasonably good closure. Therefore, in addition to the mapping closure, a member of the family of *Pearson* type frequencies is also considered. The results obtained by this frequency are compared with those of the mapping closure, and are also assessed against data generated by means of DNS.

In the next section, the problem under consideration is outlined along with the mathematical basis by which the single-point PDF methods are used. In Section 2.1, the salient features of the mapping closure at the single-point level are analyzed including a discussion on the formalities of the closure for our purpose. With this closure, a closed form analytical expression is provided for the limiting bound of the reactant conversion. This limiting rate is also predicted by means of the Beta density model in Section 2.2. In both these subsections, the simplifications for the case of a stoichiometric mixture are made, motivating the use of our final "simple" analytical expressions in practical modeling of stoichiometric plug flow reactors. In Section 3.1 the results predicted by both models are compared against those generated by DNS of a three-dimensional homogeneous turbulent flow. In Section 3.2 some discussions are presented highlighting the implications of these results in turbulence modeling. This paper is drawn to a close in Section 4 with some discussions for possible future extension of the two models for the statistical description of more complicated chemically reacting turbulent flows.

2 Formulation

With the assumptions described in the introduction, the statistical behavior of the reacting field in the reaction $F + rO \rightarrow (1+r)Product$ can be related to the statistics of an appropriate conserved Shvab-Zeldovich mixture fraction, $\mathcal{J}$ (Bilger, 1980). This mixture fraction can be normalized in such a way to yield values of unity in the fuel F stream and zero in the oxidizer O stream. For the purpose of statistical treatment, we define $\mathcal{P}_F(\xi, t)$, $\mathcal{P}_O(\xi, t)$ and $\mathcal{P}_{\mathcal{J}}(\xi, t)$, respectively, as the marginal PDF's of the mass fraction of F, the mass fraction of O, and the Shvab-Zeldovich variable $\mathcal{J}$. For initially segregated reactants with no fuel in the oxidizer stream (and vice versa), the initial conditions for the marginal PDF's of the mass fractions of the two reactants are given by:

$$P_F(\xi, 0) = W_F \delta(\xi - F_i) + W_O \delta(\xi),$$

$$P_O(\xi, 0) = W_O \delta(\xi - O_i) + W_F \delta(\xi), \quad 0 \leq \xi \leq 1. \tag{1}$$

Here, F_i and O_i denote the initial mass fractions of the two species in the two feeds, and W_F and W_O represent the relative weights of the reactants at the initial time (*i.e.* the area ratios at the inlet of a plug flow reactor). With the normalized value of the mass fractions equal to unity at the feeds, *i.e.* $F_i = O_i = 1$, the stoichiometric value of the Shvab-Zeldovich variable, $\mathcal{J}_{\text{st}}$ is determined from the parameter r. With the assumption of an infinitely fast chemistry, the marginal PDF's of the two reactants are related to the frequency of the Shvab-Zeldovich variable (Bilger, 1980; Kosaly and Givi, 1987):

$$\mathcal{P}_F(\xi, t) = (1 - \mathcal{J}_{\text{st}})\mathcal{P}_{\mathcal{J}}(\mathcal{J}_{\text{st}} + \xi(1 - \mathcal{J}_{\text{st}}), t) + \vartheta_F(t)\delta(\xi)$$

$$\mathcal{P}_O(\xi, t) = \mathcal{J}_{\text{st}}\mathcal{P}_{\mathcal{J}}(\mathcal{J}_{\text{st}}(1 - \xi), t) + \vartheta_O(t)\delta(\xi) \tag{2}$$

Here, $\xi \geq 0$, and

$$\vartheta_F(t) = \int_{-\infty}^{\mathcal{J}_{st}} P_{\mathcal{J}}(\xi, t) d\xi,$$

$$\vartheta_O(t) = \int_{\mathcal{J}_{st}}^{\infty} \mathcal{P}_{\mathcal{J}}(\xi, t) d\xi = 1 - \vartheta_F(t). \tag{3}$$

The initial condition for the PDF of the Shvab-Zeldovich variable is given by:

$$P_{\mathcal{J}}(\xi, 0) = W_F \delta(\xi - 1) + W_0 \delta(\xi). \tag{4}$$

Equation (4) implies that $< \mathcal{J} > (t = 0) = W_F$. Since $\mathcal{J}$ is a conserved variable, its mean value remains constant, $i.e.$ $< \mathcal{J} > (t) = < \mathcal{J} > (0) = W_F$. The integration of Eq. (2) yields the temporal variation of the statistics of the species field at all times, if the PDF's of $\mathcal{J}$ are known. As indicated above, in the setting of a mixing controlled reaction this PDF provides all the desired statistical properties of the reacting field.

2.1 Amplitude Mapping Closure

The implementation of the amplitude mapping closure involves a mapping of the random field of interest ξ to a stationary Gaussian reference field ϕ_0, via a transformation $\xi = \chi(\phi_0, t)$. Once this relation is established, the PDF of the random variable ξ, $\mathcal{P}(\xi)$, is related to that of a Gaussian distribution. In homogeneous turbulence, the transport equation for this function satisfies (Chen et $al.$, 1989; Pope, 1991):

$$\frac{\partial \chi}{\partial \tau} = -\phi_0 \frac{\partial \chi}{\partial \phi_0} + \frac{\partial^2 \chi}{\partial \phi_0^2}. \tag{5}$$

In this equation, τ is a normalized time within which the scalar length scale information

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is imbedded. The relations between this time and the physical time, *i.e.* $\tau(t)$, cannot be determined in the context of single-point PDF description and must be provided by external means (Pope, 1990; Jiang *et al.*, 1992). For the case considered here, with the initial PDF of the variable $\mathcal{J}$ given by Eq. (3), the corresponding form of the initial mapping is:

$$\chi(\phi_0, 0) = H(\phi_0 - \phi^*), \quad -\infty \le \phi_0 \le \infty, \tag{6}$$

where H is the Heaviside function, and ϕ^* is a measure of the initial asymmetry of the initial PDF around the ensemble mean of the variable $\mathcal{J}$:

$$\phi^* = \sqrt{2}\text{erf}^{-1}(1 - 2 < \mathcal{J} >), \tag{7}$$

where "erf" denotes the error function. The mapping function is obtained by solving Eq. (5) subject to initial condition (6). The general solution of this equation has the form (Gao, 1991a):

$$\chi(\phi_0, \tau) = \frac{\sqrt{\mathcal{G}^2 + 1}}{\sqrt{2\pi}\mathcal{G}} \int_{-\infty}^{\infty} \chi(y, 0) \exp\left[-\frac{(\phi_0 e^{-\tau} - y)^2 (1 + \mathcal{G}^2)}{2\mathcal{G}^2}\right] dy. \tag{8}$$

where $\mathcal{G}(\tau) = \sqrt{\exp(2\tau) - 1}$. Inserting Eq. (6) for $\chi(\phi_0, 0)$ in Eq. (8), we have:

$$\chi(\phi_0, \tau) = \frac{1}{2}\left[1 + \text{erf}(a\phi_0 + b)\right], \tag{9}$$

where,

$$a(\tau) = \frac{1}{\sqrt{2}\mathcal{G}}, \quad b(\tau) = \frac{-\phi^*\sqrt{1 + \mathcal{G}^2}}{\sqrt{2}\mathcal{G}}. \tag{10}$$

Finally, the solution for the PDF of $\mathcal{J}$ is determined directly from the mapping relation between the physical field ξ and the Gaussian reference field ϕ_0:

$$\mathcal{P}_{\mathcal{J}}\left(\chi(\phi_0,\tau),\tau\right) = P_G(\phi_0)\left(\frac{\partial \chi}{\partial \phi_0}\right)^{-1}. \tag{11}$$

Here, P_G denotes the PDF of a standardized Gaussian distribution, *i.e.*, $P_G(\phi_0) = \frac{1}{\sqrt{2\pi}}\exp(-\phi_0^2/2)$. A combination of Eq. (11) and Eq. (9), yields the final result for PDF of the Shvab-Zeldovich variable:

$$\mathcal{P}_{\mathcal{J}}\left(\chi(\phi_0,\tau),\tau\right) = \mathcal{G}\exp\left[\frac{(\phi_0 e^{-\tau} - \phi^*)^2}{2(1 - e^{-2\tau})} - \frac{\phi_0^2}{2}\right]. \tag{12}$$

With a combination of Eqs. (12) and (2), all the pertinent single-point statistics of the reacting field are determined. The most important of these statistics are the ensemble mean values of the reactants' concentrations. These mean values are obtained directly by integrating their respective PDF's. The intermediate steps in deriving these relations are not presented but are provided by Frankel (1992). Here, only the essential steps are presented. For the mean fuel concentration, $< F >$ the first part of Eq. (2) reads:

$$< F > (\tau) = \int_{\mathcal{J}_{st}}^{1} F(\xi)\mathcal{P}_{\mathcal{J}}(\xi,\tau)d\xi = \int_{\phi_0(\chi=\mathcal{J}_{st})}^{\infty} F(\chi,\tau)P_G(\phi_0)d\phi_0, \tag{13}$$

where the lower limit of the last integral corresponds to the value of ϕ_0 at which χ is equal to the stoichiometric value of the Shvab-Zeldovich variable. Evaluating this limit from Eq. (9), Eq. (13) can be analytically integrated. This is possible by representing the error function in the form of its definition, and performing the resulting definite, double exponential integral. The results after extensive algebraic manipulations yield:

$$< F > (\tau) = \frac{(1 - 2\mathcal{J}_{st})}{4(1 - \mathcal{J}_{st})}\left[1 + \text{erf}\left(\frac{b/a + c}{\sqrt{2}}\right)\right] + \frac{1}{\pi\sqrt{2}a(1 - \mathcal{J}_{st})}\exp\left(-\frac{b^2}{2a^2} - \frac{c^2}{2} - \frac{bc}{a}\right)\Theta_+ \tag{14}$$

Similar expressions can be obtained for the mean oxidizer concentration:

$$< O > (\tau) = \frac{1}{2}(1 - \frac{1}{2\mathcal{J}_{\text{st}}}) \left[1 - \text{erf}\left(\frac{b/a + c}{\sqrt{2}} \right) \right] - \frac{1}{\pi\sqrt{2}a\mathcal{J}_{\text{st}}} \exp\left(-\frac{b^2}{2a^2} - \frac{c^2}{2} - \frac{bc}{a} \right) \Theta_- \quad (15)$$

In these equations, c is related to the stoichiometric coefficient,

$$c = \frac{-1}{a}\text{erf}^{-1}(2\mathcal{J}_{\text{st}} - 1) \quad (16)$$

and

$$\Theta_\pm = \int_0^1 dy \exp\left(-a^2c^2y^2 + \frac{y_\pm^2}{8y_2} \right) \left[\frac{1}{2y_2}\mathcal{D}_{-2}\left(\frac{y_\pm}{\sqrt{2y_2}} \right) - \frac{ac}{\sqrt{2y_2}}\mathcal{D}_{-1}\left(\frac{y_\pm}{\sqrt{2y_2}} \right) \right],$$

$$y_2 = y^2 + \frac{1}{2a^2},$$

$$y_\pm = \mp 2acy^2 - \frac{b}{a^2} - \frac{c}{a}. \quad (17)$$

Here, $\mathcal{D}_{-2}$ and $\mathcal{D}_{-1}$ are, respectively, the Parabolic Cylinder Functions of order -2 and -1, belonging to the family of degenerate hypergeometric functions (Abramowitz and Stegun, 1972).

Due to nice properties of the degenerate hypergeometric functions, many of the interesting features of Eqs. (14)-(15) can be depicted. First, simple manipulation of this equation shows that for a stoichiometric mixture, $< \mathcal{J} > = \mathcal{J}_{\text{st}}$, both reactants decay at the same rate, *i.e.* $\frac{<F>(t)}{<F>(0)=W_F} = \frac{<O>(t)}{<O>(0)=W_O}$, and for a non-stoichiometric initial condition, the limits of the mass fractions as $\tau, \mathcal{G} \to \infty$, asymptote to:

$$\text{Lim}_{\tau,\mathcal{G}\to\infty} < F > (\tau) = \begin{cases} 0 & \mathcal{J}_{\text{st}} \geq < \mathcal{J} > \\ \frac{<\mathcal{J}>-\mathcal{J}_{\text{st}}}{1-\mathcal{J}_{\text{st}}} & \mathcal{J}_{\text{st}} \leq < \mathcal{J} > \end{cases}$$

$$\text{Lim}_{\tau, \mathcal{G} \to \infty} < O > (\tau) = \begin{cases} 0 & \mathcal{J}_{st} \leq <\mathcal{J}> \\ 1 - \frac{<\mathcal{J}>}{\mathcal{J}_{st}} & \mathcal{J}_{st} \geq <\mathcal{J}> \end{cases} \qquad (18)$$

These limits are obtained by employing the Taylor series expansion of the relevant functions as $\mathcal{G} \to \infty$, and indicate the limiting bound of the unconsumed reactants in both fuel rich and fuel lean mixtures. While these limiting behaviors are rather trivial from a physical standpoint, in a computational procedure one must make sure that they are satisfied. Because of the mathematical properties of the Parabolic Cylinder Functions, these limiting cases can be realized in our computational procedure in a relatively easy manner. It would be very difficult to obtain these limiting behavior numerically in an integration procedure within the original unbounded domain.

At first glance, Eqs. (14)-(15) may appear somewhat complicated. However, due to nice mathematical properties of the Parabolic Cylinder Functions (Abramowitz and Stegun, 1972), these equations can be integrated rather easily within the finite domain ($0 \leq y \leq 1$). For fuel-lean or fuel-rich mixtures, the integration can be only done by means of employing numerical methods. However, for a stoichiometric mixture, the results simplify further as demonstrated below.

Stoichiometric Mixture:

For practical applications in stoichiometric plug flow reactors, the equations simplify considerably. For a stoichiometric mixture, and an initially symmetric PDF around the mean value, (*i.e.* $<\mathcal{J}> = \mathcal{J}_{st} = 1/2$), both parameters b and c are zero. Under this condition, the first terms on RHS of Eqs. (14)-(15) drop. Knowing $\mathcal{D}_{-2}(0) = \mathcal{D}_{-1}(0) = 1$, the remaining terms yield:

$$\frac{<F>(\tau)}{<F>(0)} = \frac{<O>(\tau)}{<O>(0)} = \frac{1}{\pi\sqrt{2}a} \int_0^1 \frac{dy}{2y^2 + \frac{1}{a^2}} = 1 - \frac{2\arctan\mathcal{G}(\tau)}{\pi}. \qquad (19)$$

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The simplicity of this equation is noteworthy and very pleasing. Because of this simplicity, Eq. (19) is strongly recommended for engineering predictions of the reactant conversion rate in stoichiometric homogeneous flows, such as the plug flow reactors considered in numerous previous investigations (Toor, 1962; Toor, 1975; Brodkey, 1981; Hill, 1976; O'Brien, 1971; Kosaly and Givi, 1987).

2.2   Beta Density Model

For initially segregated reactants, the initial PDF of the Shvab-Zeldovich variable is composed of two delta functions at the extreme limits of the variable. Therefore, it is proposed that the family of Pearson (1895) frequencies may provide a reasonable means of estimating this distribution at all times (Frankel *et al.*, 1991; Girimaji, 1991a). The appropriate form of the Pearson distribution is in this case the Beta density of the first kind. This density has been employed in the statistical description of turbulent reacting flows by (Rhodes, 1975; Jones and Priddin, 1978; Lockwood and Moneib, 1980; Peters, 1984; Janicka and Peters, 1982) amongst others (for recent reviews, see (Givi, 1989; Priddin, 1991)). For an initially non-symmetric PDF, the Beta density corresponds to the Pearson Type I and for the symmetric case to Pearson Type II. The relevance of the latter in modeling of molecular mixing from an initial symmetric binary state has been described by Madnia *et al.* (1991a); Girimaji (1991a). In both cases, the PDF of the Shvab-Zeldovich variable is represented by (Abramowitz and Stegun, 1972):

$$\mathcal{P}_{\mathcal{J}}(\xi) = \frac{1}{B(\beta_1, \beta_2)} \xi^{\beta_1 - 1} (1 - \xi)^{\beta_2 - 1}, \ 0 \leq \xi \leq 1. \tag{20}$$

where $B(\beta_1, \beta_2)$ denotes the Beta function, and the parameters β_1 and β_2 are dependent on the mean and the variance of the random variable $\mathcal{J}$. In applications to the mixing controlled reaction considered here, we assume that the PDF of the Shvab-Zeldovich variable always retains a Beta distribution. Thus all the statistics of the reacting scalar are subsequently determined. The ensemble mean values are determined by a combination of Eqs. (20) and

(2). Following the same procedure as that described in the section on the mapping closure, after some manipulations the final results can be expressed as (Frankel, 1992):

$$< F > (t) = \frac{\mathcal{J}_{st}^{\beta_1}(1 - \mathcal{J}_{st})^{\beta_2 - 1}}{(\beta_1 + \beta_2)B(\beta_1, \beta_2)} + \frac{1}{1 - \mathcal{J}_{st}} \left(\frac{\beta_1}{\beta_1 + \beta_2} - \mathcal{J}_{st} \right) (1 - \mathcal{I}_{\mathcal{J}_{st}}(\beta_1, \beta_2)) \qquad (21)$$

$$< O > (t) = \frac{\mathcal{J}_{st}^{\beta_1 - 1}(1 - \mathcal{J}_{st})^{\beta_2}}{(\beta_1 + \beta_2)B(\beta_1, \beta_2)} + \left(1 - \frac{\beta_1}{(\beta_1 + \beta_2)\mathcal{J}_{st}} \right) \mathcal{I}_{\mathcal{J}_{st}}(\beta_1, \beta_2), \qquad (22)$$

where $\mathcal{I}$ denotes the Incomplete Beta Function (Abramowitz and Stegun, 1972).

Due to nice mathematical properties of the Beta Function, the final results are cast in terms of its integral. In this case, however, the integral can be expressed in terms of the known special Incomplete Beta function. The mathematical properties of this function are well known, and the expressions above are conveniently amenable to numerical integration (Frankel, 1992). Again, the physical limiting conditions discussed before are realized by Eqs. (21)-(22). That is, in a stoichiometric mixture, both reactants decay at the same rate; and in lean or rich mixtures, the same limiting conditions as those in Eq. (18) are realized.

Stoichiometric Mixture

Again, in the case of an initially symmetric PDF under stoichiometric conditions, the final expressions become simpler. Under this condition, $\beta_1 = \beta_2$, and knowing $\mathcal{I}_{1/2}(x, x) = 1/2$, Eqs. (21)-(22) reduce to:

$$\frac{< F > (t)}{< F > (0)} = \frac{< O > (t)}{< O > (0)} = \frac{1}{\sqrt{\pi}} \frac{\Gamma(g)}{\Gamma(g + \frac{1}{2})}, \qquad (23)$$

where g is half the inverse of the normalized variance of the Shvab-Zeldovich variable, and Γ denotes the Gamma function.

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3 Results

The final forms of Eqs. (14), (15), (19), (21), (22), and (23) are gratifying since they provide a relatively simple and effective means of estimating the maximum rate of reactant conversion in homogeneous reacting flows. As indicated before, the mathematical operations leading to these equations are somewhat involved, but the final results can be conveniently expressed in terms of known special functions. However, since both models are based on single-point PDF descriptions, these equations are not in a complete closed form and are dependent on the parameters g and/or $\mathcal{G}$. In this context, this parameter cannot be determined by the model and must therefore be specified by external means (Pope, 1991; Jiang *et al.*, 1992; Frankel *et al.*, 1992). This deficiency is not particular to the two models considered here, and exists in any single-point statistical description, including all of those based on the C/D models.

The extent of validity of these simple relations can be demonstrated by a comparison between the model predictions and the data obtained by means of DNS. The comparison is made here for several values of $< \mathcal{J} >$ and $\mathcal{J}_{st}$ for the purpose of demonstration. In this comparison, the magnitudes of the normalized variance of the Shvab-Zeldovich variable are matched with those of DNS. This implies that for given values of $< \mathcal{J} >$ and $\mathcal{J}_{st}$, the parameters $g, \mathcal{G}, \beta_1$, and β_2 are provided externally from the DNS data. With this provision, the model prediction results can be directly assessed against DNS data.

The DNS procedure is similar to that of previous simulations of this type. For a detailed description, we refer the reader to Madnia and Givi (1992). The subject of the present DNS is a three-dimensional periodic homogeneous box flow under the influence of a binary reaction of the type described above. The initial species field is assumed to be composed of out of phase square waves for the two reactants F and O. The computational package is based on the modification of a spectral-collocation procedure using Fourier basis functions developed by Erlebacher *et al.* (1990a) (also see Erlebacher *et al.* (1987); Erlebacher *et al.* (1990b)). The hydrodynamic field is assumed isotropic, and is initialized in a similar manner to that of Erlebacher *et al.* (1990a); Passot and Pouquet (1987). The turbulent field is of decaying

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nature in that there is no artificial forcing mechanism to feed energy to low wave numbers. The code is capable of simulating flows with different levels of compressibility (Hussaini *et al.*, 1990). Here, only the results obtained for a low compressible case are discussed, since most previous analyses of plug flow reactors have dealt primarily with incompressible flows (Toor, 1975; Hill, 1976; Brodkey, 1981; Leonard and Hill, 1988b; Leonard and Hill, 1991). The resolution consists of 96 collocation points in each direction. Therefore, at each time step 96^3 is the sample size for statistical analyses. With this resolution, simulations with a Reynolds number (based on the Taylor microscale) of $Re_\lambda \approx 41$ are attainable. The value of the molecular Schmidt number is set equal to unity.

3.1   Validations

The statistical behavior of the scalar field is depicted by examining the evolution of the PDF's of the Shvab-Zeldovich variable $\mathcal{J}$. These are shown in Figs. 1 and 2 at times close to the initial ($t1$) and the final ($t2$) states. These figures correspond, respectively, to the cases of an initially symmetric ($< \mathcal{J} >= 1/2$) and non-symmetric ($< \mathcal{J} >= 0.625$) PDF's. At the initial time, the PDF is approximately composed of two delta functions at $\mathcal{J} = 0, 1$ indicative of the two initially segregated reactants, F and O. At later times, the PDF evolves through an inverse-like diffusion in composition space. The heights of the delta functions decrease, and the PDF is redistributed at other $\mathcal{J}$ values in the range $[0, 1]$, and subsequently becomes centralized around the mean value. Proceeding further in time results in a sharper peak at this mean value, and in both cases, the PDF can be approximated by a Gaussian distribution. The trend for the symmetric case is the same as that presented in earlier DNS studies (Eswaran and Pope, 1988; Givi and McMurtry, 1988). For the non-symmetric case, there are no DNS data in the literature, but the present results verify that the asymptotic PDF can still be approximated by a Gaussian distribution.

The PDF's obtained by the mapping closure and those assumed by a Beta density are also presented in Figs. 1-2. In these figures, the model PDF's are parameterized with the same first two moments obtained from DNS. In this parameterization, only the normalized

magnitude of the variance of the models are forced equal to that of the DNS and no attempt was made to account for the departure from the "exact" initial double delta distribution in DNS. With this matching, nevertheless, the results clearly indicate that the model predictions compare very well with the DNS results. Also, both models yield an asymptotic Gaussian PDF.

The temporal variation of the ensemble mean of the reactants' mass fractions by the two models are compared against those of DNS in Figs. 3-4. These figures correspond to the two cases of symmetric and non-symmetric initial PDF's, respectively. In the symmetric case, under stoichiometric conditions, the results are simply obtained from the analytical expressions in Eqs. (19) and (23). In the non-symmetric case, the numerical integration of Eqs. (14)-(15), and the evaluation of the Incomplete Beta Function in Eqs. (21)-(22) are necessary. In both cases, the agreement of the models with the DNS data is noteworthy. Also, a comparison between parts (a) and (b) of Fig. 4 shows that as the magnitude of $\mathcal{J}_{st}$ decreases, the rate of oxidizer consumption becomes faster.

The agreement noted above is pleasing but not very surprising. It follows from the compatibility of the model PDF's and those of DNS, at least for the case of binary mixing considered here. This finding is not new and has been well documented in previous works, at least those considering an initially symmetric PDF (Pope, 1990; Pope, 1991; Madnia and Givi, 1992; Madnia *et al.*, 1991a; Girimaji, 1991a). However, a nice feature of the models is the explicit form of the final equations expressing these statistical quantities. Supported by this quantitative agreement, it is proposed that in the absence of a better alternative, the relations obtained above to be used as an explicit simple means for predicting the maximum rate of reactant conversion in homogeneous reacting systems.

Despite the simplicity of these equations and their ease of applications, it must be mentioned that these equations predict the rate of mean decay of the reactant far better than all of the previous turbulence closures based on the C/D models (see Givi (1989)). This is particularly advantageous in that this evaluation can be made by a simple algebraic relation, whereas the C/D implementations usually require more expensive numerical simulations. Even for

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non-unity equivalence ratios, the numerical integration required by the two above models is considerably less computationally demanding than those of the C/D models. The only input in these models, similar to those in C/D closures, is the variance of the Shvab-Zeldovich variable. This is provided here by means of DNS. In an actual implementation, this variance can be obtained from experimental data or by means of an appropriate turbulence model (Frankel *et al.*, 1992). The provision of such data is not very difficult since they can be obtained in the setting of a non-reacting flow.

3.2 Applications

The relations obtained here can be used in determining the extent of validity of other conventional closures for predicting the limiting rate of reactant conversion in turbulent flows. As an example, here we consider the model based on the famous hypothesis of Toor (1962); Toor (1975), which has received considerable attention in practical modeling of unpremixed homogeneous reacting systems (Bilger, 1980; Brodkey, 1981; Leonard and Hill, 1987; Leonard and Hill, 1988a; Leonard and Hill, 1988b; Kosaly and Givi, 1987; Kosaly, 1987; Givi and McMurtry, 1988; McMurtry and Givi, 1989; Givi, 1989) . According to this hypothesis, in an isothermal homogeneous reacting turbulent flow, the decay of the unmixedness denoted by $< a'b' >$, where the prime quantities indicate fluctuation from the ensemble mean value, is independent of the magnitude of the Damköhler number. This implies that the normalized unmixedness parameter, defined by:

$$\mathcal{Z} = \frac{< a'b' > (t)|_{Da}}{< a'b' > (t)|_{Da=0}}, \tag{24}$$

is the same under both reacting and non-reacting conditions, *i.e.* $\mathcal{Z}(t) = \text{constant} = 1$ for all values of Da. In previous DNS assessments of this hypothesis, it has been shown that for the case of initially segregated reactants this model cannot be employed, and the normalized unmixedness ratio depends on the nature of mixing and the magnitude of the Damköher number (Givi and McMurtry, 1988; McMurtry and Givi, 1989). In particular, it has been

demonstrated that even for $Da \to \infty$, while the normalized unmixedness is equal to unity at the initial time, its limiting lower bound depends on the asymptotic frequency of the Shvab-Zeldovich variable. That is, $\mathcal{Z}(t=0) = 1 > \mathcal{Z}(t) > \mathcal{Z}(t \to \infty) = C$, where C is the lower limiting bound. For an asymptotic Gaussian distribution, it can be easily shown that for a mixture under stoichiometric conditions, the lower bound limit asymptotes to the constant value $C = 2/\pi$ (Kosaly, 1987; Givi and McMurtry, 1988).

This deviation from unity can be realized by the two models considered. With the mapping closure, under symmetric stoichiometric conditions, from Eqs. (2),(14), and (15), following the same integration procedure as before, it is shown that:

$$\mathcal{Z}_m = \frac{< a'b' > (Da \to \infty)}{< a'b' > (Da = 0)} = \frac{2 \left(\arctan(\frac{1}{\mathcal{G}}) \right)^2}{\pi \arctan \left(\frac{1}{\mathcal{G}\sqrt{\mathcal{G}^2+2}} \right)}. \tag{25}$$

For the Beta density model, the corresponding form of the unmixedness ratio for $\beta_1 = \beta_2$ is given by:

$$\mathcal{Z}_\beta = \frac{2g}{\pi} \left(\frac{\Gamma(g)}{\Gamma(g + \frac{1}{2})} \right)^2. \tag{26}$$

In Eqs. (25)-(26), the subscripts m, β are added to denote the mapping and the Beta density model. It is easy to show that these equations satisfy the correct limiting conditions for a stoichiometric mixture. This is for both the initial time, *i.e.* the inlet of the reactor,

$$\text{Lim}_{[\mathcal{G} \to 0]} \mathcal{Z}_m = \text{Lim}_{[g \to \frac{1}{2}]} \mathcal{Z}_\beta = 1, \tag{27}$$

and at large distances form it:

$$\text{Lim}_{[\mathcal{G} \to \infty]} \mathcal{Z}_m = \text{Lim}_{[g \to \infty]} \mathcal{Z}_\beta = C = \frac{2}{\pi}. \tag{28}$$

The latter limiting condition cannot be realized in any of the previously used C/D models, or by means of the Toor's models (McMurtry and Givi, 1989; Givi, 1989).

The results based on the applications of Toor's model become less accurate for non-stoichiometric mixtures. For equivalence ratios other than unity, with the depletion of one of the reactants, the unmixedness parameter approaches zero faster. This is demonstrated in Fig. 5 for several values of the equivalence ratio (γ). Note that as the magnitude of this ratio increases above one, the unmixedness ratio goes to zero more rapidly. For an unity equivalence ratio, the correct asymptotic value of $2/\pi$ is realized.

4 Extensions for Modeling of More Complex Reacting Turbulent Flows

Despite the pleasing features of our simple mathematical expressions, there are several restricting assumptions which were necessarily imposed in deriving these equations. Here, we would like to address the ramifications associated with these assumptions, and to provide the means of overcoming them in future extensions of these two models.

Firstly, due to the assumption of infinitely fast chemistry, only the *maximum rate* of the reactant conversion is obtained. While this rate is of crucial interest in describing unpremixed flames, from both a theoretical standpoint and for practical applications (Givi, 1989; McMurtry and Givi, 1989; Kosaly and Givi, 1987; Toor, 1962; Toor, 1975; O'Brien, 1971; Bilger, 1980; Williams, 1985), the model is not capable of describing some of the important features of the turbulent flames, especially those under non-equilibrium conditions. The extensions to finite rate chemistry, reversible reactions, non-equilibrium flames and multi-step kinetics systems require numerical integration of the PDF transport equation. For these cases, the problem cannot be mathematically reduced to that of keeping track of a single scalar variable (like $\mathcal{J}$), and requires the use of *multivariate* statistical descriptions. For this, the implementation of mapping closure is relatively straightforward since it provides a trans-

port equation for the joint PDF's of the scalar variable in a multivariable sense (Pope, 1991; Gao and O'Brien, 1991). However, it is not presently clear how to devise an efficient computational procedure, typically based on Monte Carlo methods (Pope, 1981), for the numerical treatment of these equations. Some work in this regard is currently under way (Valiño and Gao, 1991; Valiño et al., 1991).

The extension of assumed distributions based on the Beta density for treating multi-scalars is more straightforward but less trivial to justify. The most obvious means is to implement the multivariate form of the Pearson distributions. The direct analogue of the Beta density is the *Dirichlet* frequency (Johnson, 1987; Narumi, 1923) . For a mixture composed of $N+1$ species, the joint PDF of N mass fractions $(\psi_1, \psi_2, \ldots \psi_N)$ is described in terms of a N-variate density of the form:

$$\mathcal{P}(\psi_1, \psi_2, \ldots, \psi_N) = \frac{\Gamma(\beta_1 + \beta_2 + \ldots \beta_{N+1})}{\Gamma(\beta_1)\Gamma(\beta_2), \ldots \Gamma(\beta_{N+1})} \psi_1^{\beta_1-1} \psi_2^{\beta_2-1} \ldots \psi_N^{\beta_N-1} (1-\psi_1-\psi_2-\ldots\psi_N)^{\beta_{N+1}-1}$$

(29)

subject to the physical constraint:

$$\sum_{i=1}^{N+1} \psi_i = 1 \tag{30}$$

The application of this density in modeling of multi-species reactions has been nicely discussed by Girimaji (1991a). Due to the mathematical properties of the Gamma function, this density is pleasing from a mathematical viewpoint and most statistical cross correlations of the random variables $(\psi_1, \psi_2, \ldots)$ can be conveniently obtained by means of simple analytical relations (Frankel, 1992). Some points in this regard has been made by Girimaji (1991b). However, the use of the Dirichlet frequency cannot be justified for modeling of unpremixed reacting flow in a general sense. In fact, it can be shown that in the case of a binary irreversible reaction of the type considered here, if the joint PDF of the two species can be assumed to be of a bivariate Dirichlet then the univariate PDF of the Shvab-Zeldovich vari-

able cannot be approximated by a Beta density. Moreover, there is no way of implementing this density directly for modeling of non-equilibrium flames, involving strong temperature variations. This is simply due to the additivity constraints of this density requiring the unity sum of the normalized random variables (Eq. 30).

Secondly, the mathematical derivations presented here are only valid for initially segregated reactants. In both models, the complete segregation facilitates significant simplifications of the final equations. This assumption is not unrealistic and is compatible with that in majority of previous works on unpremixed reacting flows (Toor, 1975; Brodkey, 1981; Bilger, 1980). For other initial conditions, *e.g.* partial premixing of the reactants, or non-delta like distributions, one must resort to numerical integration of the PDF transport equation. Again, an appropriately devised numerical procedure can accommodate such conditions. But the use of a Beta density (or any other assumed distributions) cannot be justified for other complex initial conditions.

Thirdly, the final mathematical expressions are only valid in the setting of a homogeneous flow. The extension to inhomogeneous flow predictions is also straightforward, but requires numerical integration of the modelled equations. Both models can be directly implemented into appropriately devised numerical procedures. The mapping closure can be invoked in the *mixing* step of a fractional stepping procedure, similar to that of typical Monte Carlo procedures (Pope, 1981). The Beta density requires modelled transport equations for the low order statistics of the reacting field. These equations include the required information pertaining to the spatial inhomogeneity of the flow, through the parameters $\beta_1, \beta_2, \ldots$. With these informations, all the higher order statistics of the reacting field can be provided by simple analytical means (Girimaji, 1991b).

Finally, in the context of single-point PDF formulation presented, there is no information pertaining to the evolution of the relevant turbulent length scales. The final expressions can be only presented in terms of other physical parameters (here, only through the parameters g and/or $\mathcal{G}$). In the context considered, this parameter has been provided by the DNS data. In a practical application, this must be provided by external means (turbulence models,

experimental data, *etc.*) Jiang *et al.* (1992), and Frankel *et al.* (1992) provide further discussions related to this issue.

5   Concluding Remarks

It is demonstrated that the mapping closure of Kraichnan-Pope (Chen *et al.*, 1989; Pope, 1991) yields closed form analytical expressions for predicting the limiting bound of reactant conversion rate in a simple chemistry of the type $F + rO \rightarrow (1 + r)$ *Product* in homogeneous, isothermal turbulence. It is also shown that for the case of complete initial segregation, the scalar PDF's generated by this closure can be well-approximated by a Beta density. This density also provides closed form analytical expressions for the limiting rate of reactant conversion. A nice feature of the mathematical results generated by the two models is their capability of revealing the influence of the stoichiometric coefficient and the equivalence ratio. In both cases, the mathematical expressions simplify significantly for a stoichiometric mixture. The prediction results via both models compare favorably with data generated by DNS. This agreement is not surprising, since for the case of a mixing controlled reaction, it follows from the compatibility of the models PDF's with those of DNS. However, the simple final results generated here are superior to those of previous closures based on the typical C/D models, and those based on Toor's hypothesis. For example, in comparison with Toor's results, it is shown that in stoichiometric mixtures, an improvement of approximately 36% can be made in evaluating the unmixedness; for non-stoichiometric mixtures the improvement can be as high of 100%, depending on the value of the equivalence ratio.

These closed form relations are furnished with the imposition of several restrictive assumptions. The ramifications associated with these assumptions are discussed, and some suggestions for future extensions are provided. In spite of these assumptions, it is very encouraging to have physically plausible algebraic relations for direct estimate of the reactant conversion rate in plug flow reactors. Because of their simplicity and demonstrated validity, these expressions are strongly recommended for routine and economical engineering predictions in

unpremixed binary reacting systems such as those in plug flow reactors.

Nomenclature

a, b, c. some constant.

B. the Beta function.

$\mathcal{D}$. the Parabolic Cylinder Function.

Da. Damköhler number.

F. Fuel.

$\mathcal{G}$. parameter in the mapping closure.

g. half the inverse of the normalized variance of the Shvab-Zeldovich variable.

H. the Heaviside function.

$\mathcal{I}$. the incomplete Beta function.

$\mathcal{J}$. Shvab-Zeldovich variable.

O. Oxidizer.

$\mathcal{P}$. PDF.

r. the stoichiometric coefficient.

t. physical time

W. area weight of the reactant.

y. dummy variable of integration.

$\mathcal{Z}$. the unmixedness ratio.

Greek letters

$\beta_1, \beta_2, \ldots$. parameters of the Beta density.

γ. equivalence ratio.

Γ. the Gamma Function

δ. the delta function.

ξ. the composition space for the PDF.

ϕ_0. composition space for a Gaussian reference field.

τ. normalized time.

χ. the mapping function.

Subscripts

0. time zero (inlet of plug flow reactor).

G. Gaussian.

st. stoichiometric.

Other symbols

$\langle \rangle$. Probability Average.

Figure Captions

Figure 1: PDF of the Shvab-Zeldovich variable at two times ($t2 > t1$) for the symmetric case ($< \mathcal{J} >= \frac{1}{2}$).

Figure 2: PDF of the Shvab-Zeldovich variable at two times ($t2 > t1$) for the non-symmetric case ($< \mathcal{J} >= 0.625$).

Figure 3: Normalized mean concentration of fuel and the oxidizer for the symmetric case, and $\mathcal{J}_{st} = 0.5$.

Figure 4: Normalized mean concentration of fuel and the oxidizer for the non-symmetric case. (a) $\mathcal{J}_{st} = 0.4$, (b) $\mathcal{J}_{st} = 0.2$.

Figure 5: The unmixedness ratio at several values of the equivalence ratio.

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FIG. # 1

FIG. #2
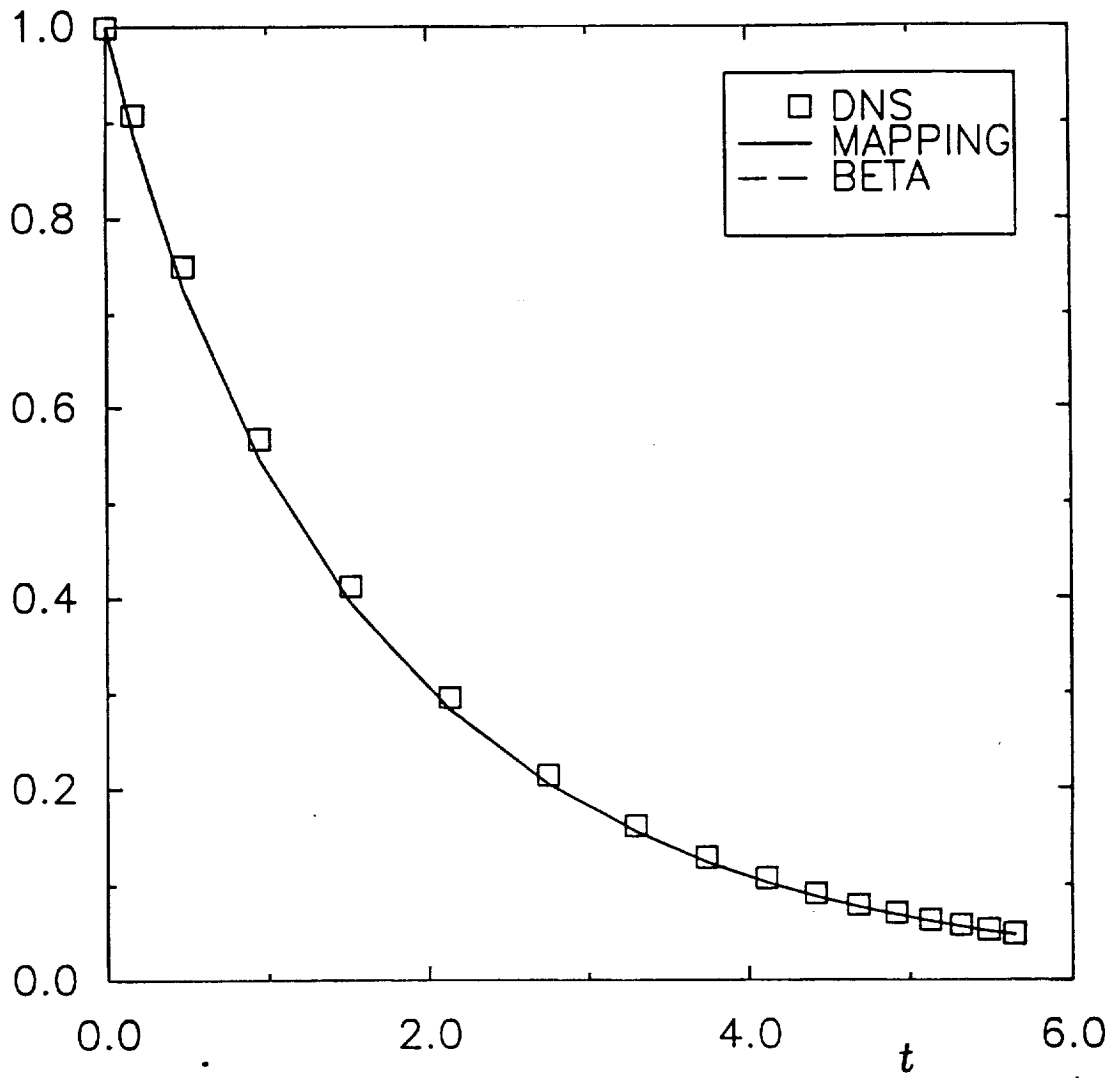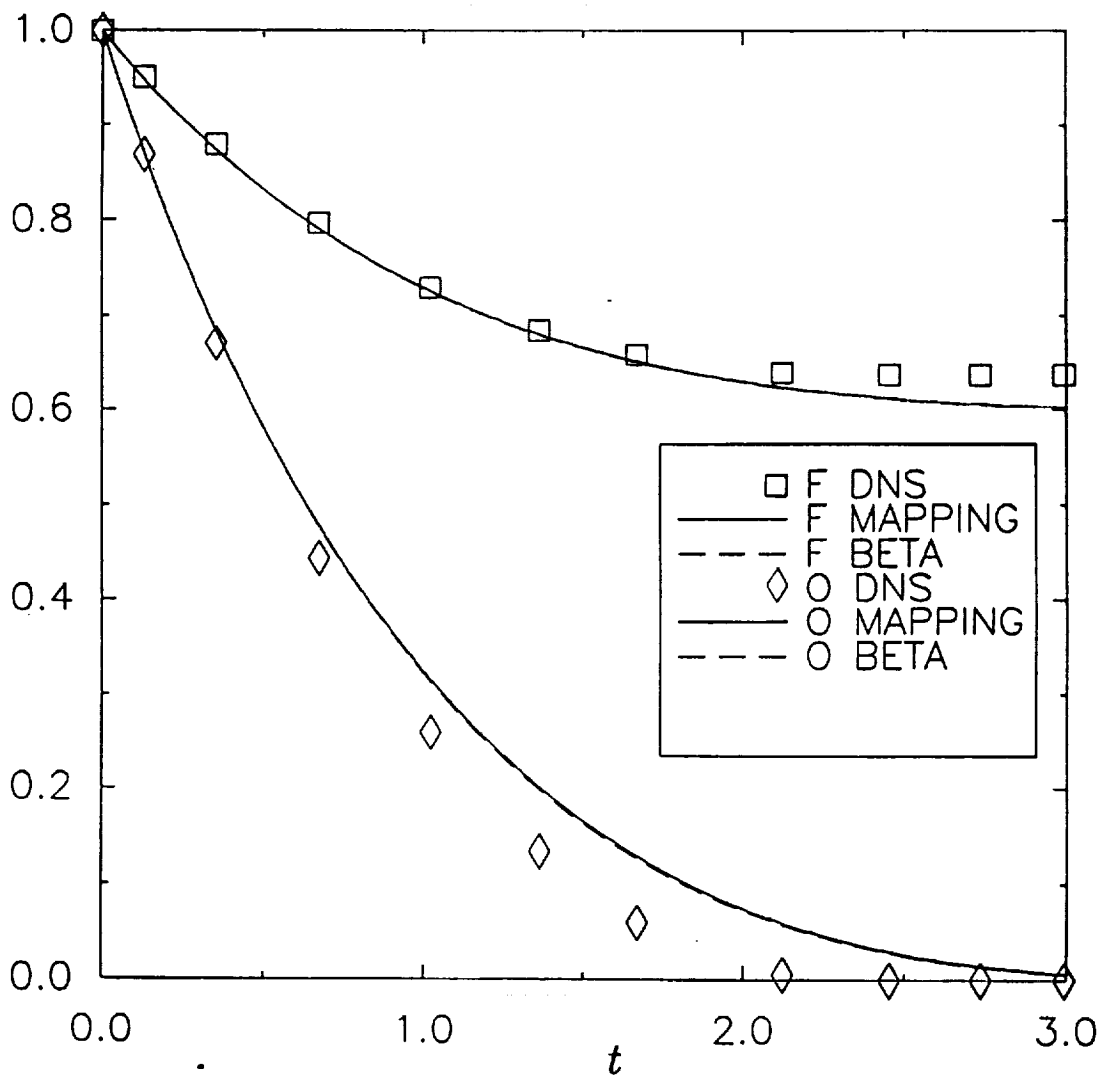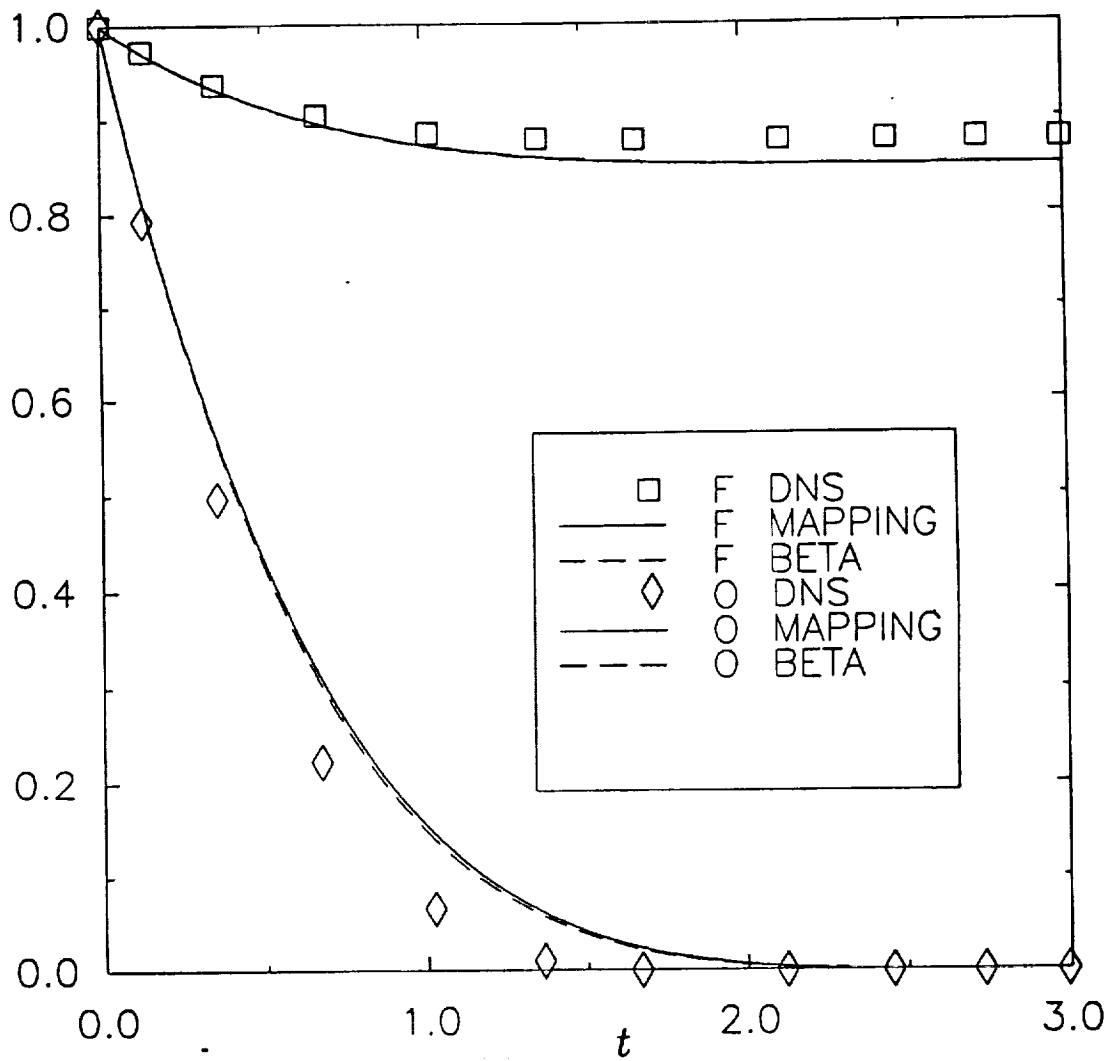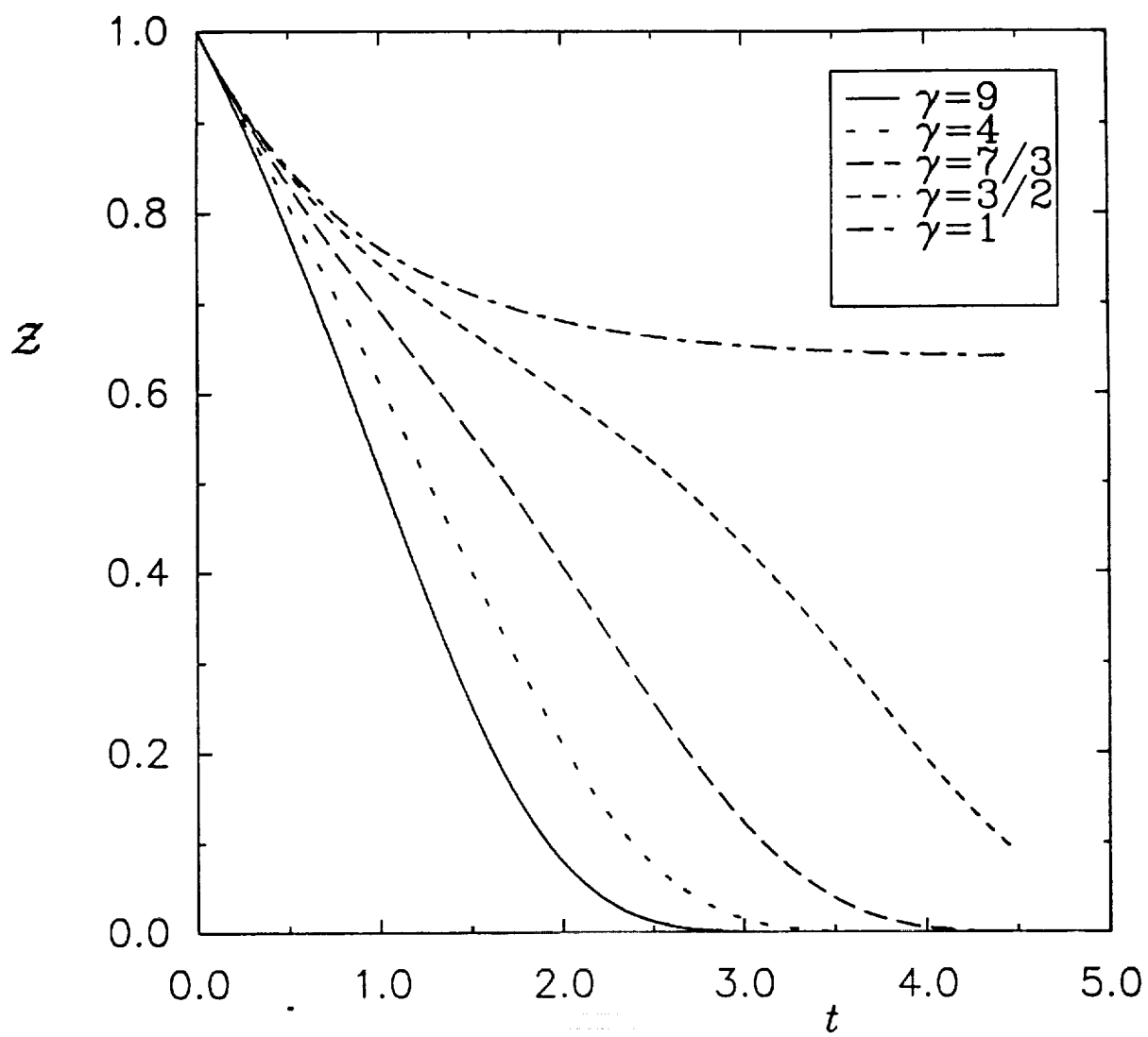
$$\frac{<F>(t)}{<F>(0)}$$

$$\frac{<O>(t)}{<O>(0)}$$

1.0

0.8

0.6

0.4

0.2

0.0

□ DNS
—— MAPPING
– – BETA

0.0   2.0   4.0   6.0

t

FIG. #3

$$\frac{<F>(t)}{<F>(0)}$$

$$\frac{<O>(t)}{<O>(0)}$$

□	F DNS
——	F MAPPING
– –	F BETA
◇	O DNS
——	O MAPPING
– –	O BETA

t

FIG. # 4(a)

$$\frac{<F>(t)}{<F>(0)}$$

$$\frac{<O>(t)}{<O>(0)}$$

	F	DNS
□	F	DNS
——	F	MAPPING
– –	F	BETA
◇	O	DNS
——	O	MAPPING
– –	O	BETA

FIG. # (4b)

$\gamma=9$
$\gamma=4$
$\gamma=7/3$
$\gamma=3/2$
$\gamma=1$

FIG. # 5

MODELING OF THE REACTANT CONVERSION RATE IN A TURBULENT SHEAR FLOW

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Results are presented of direct numerical simulations (DNS) of a spatially developing shear flow under the influence of an infinitely fast chemical reactions of the type $A + B \rightarrow Products$. The simulation results are used to construct the compositional structure of the scalar field in a statistical manner. The results of this statistical analysis indicate that the use of a Beta density for the probability density function (PDF) of an appropriate Shvab–Zeldovich mixture fraction provides a very good estimate of the limiting bounds of the reactant conversion rate within the shear layer. This provides a strong justification for the implementation of this density in practical modeling of non-homogeneous turbulent reacting flows. However, the validity of the model cannot be generalized for predictions of higher order statistical quantities. A closed form analytical expression is presented for predicting the maximum rate of reactant conversion in non-homogeneous reacting turbulence.

INTRODUCTION

In our previous work (Madnia *et al.*, 1991a) we have demonstrated that the Beta density provides a very good means of approximating the probability distribution of a conserved scalar quantity in homogeneous turbulent flows. The versatility of this density enables it to predict the evolution of a conserved scalar variable in a statistically homogeneous field, in a manner that is in accord with both DNS and experimental measurements. Despite this positive feature, some questions pertaining to the generality of this density for more realistic flow configurations remained unanswered.

  In this work, we intend to address some of these remaining questions. Specifically, we shall make a comparative assessment of the behavior of this model, and provide an estimate of the degree of its validity in the context of more general conditions than previously considered. For this purpose, we have selected a configuration of particular interest in the design of diffusion controlled reactors; namely, a spatially developing parallel mixing layer under the influence of harmonic perturbations. The flow field which develops in this setting is dominated by large scale coherent structures and exhibits non-homogeneous features. This

inhomogeneity plays a dominant role in the mechanism of mixing in turbulent flows and is very suitable for promoting mixing and chemical reactions. Because of this property, this configuration has been widely utilized in numerous investigations on turbulent reacting flow phenomena (see Givi and Riley (1992) for a recent review).

We shall follow an analogous approach to that of our previous work (Madnia *et al.*, 1991a) in assessing the model's behavior. However, there are fundamental differences between the works in formulating the problem and in implementing the numerical procedure involved in the DNS. The configuration considered here is non-homogeneous. Therefore, both the procedure of statistical sampling and the numerical algorithm used in the DNS are different from the homogeneous simulations of Madnia *et al.* Also, the mathematical operations leading to the estimate of the reactant conversion rate are substantially more elaborate. However, analytical solutions can be attained with the aid of the mathematical properties of the Beta Function and the Incomplete Beta Function. With these, the generated results can be directly compared with those of DNS.

DESCRIPTION

The problem under consideration is that of a spatially evolving mixing layer containing initially segregated reacting species A and B in the two free streams. Species A is introduced into the upper, high-speed stream and species B enters on the lower, low-speed side. The chemical reaction between the two species is assumed to be single step, irreversible and infinitely fast. The magnitude of the Mach number is assumed negligible. Therefore, the flow is considered incompressible. The two species are assumed thermodynamically similar with identical diffusion coefficients. Within the framework of these approximations, the flow is mathematically described by the conservation equations of mass, momentum, and a species conservation equation for the trace of a conserved Shvab–Zeldovich variable $\mathcal{J}$, which characterizes the compositional structure of the flow. These equations are parameterized by the Reynolds number, the Peclet number, and the velocity ratio across the layer.

The computational package employed in the DNS is based on a hybrid pseudospectral-spectral element algorithm developed by Givi and Jou (1988) and McMurtry and Givi (1991). The pseudospectral routine, which is based on Fourier collocation, is used in the cross stream direction together with free slip boundary conditions. The spectral-element discretization is employed in the streamwise direction. This involves the decomposition of the domain in this direction into an ensemble of macro finite elements. Within each of these elements, the dependent flow variables are approximated spectrally by means of Tchebysheff polynomials of the first kind. The hybrid procedure employed in this way is very attractive in that it combines the accuracy of spectral discretization with the versatility of finite element methods. Therefore, it is convenient for accurate simulations of the complex spatially developing flow under consideration

here. For a detailed description of this hybrid method for DNS of turbulent shear flows, the reader is referred to McMurtry and Givi (1991).

The flow field is initialized at the inflow by a hyperbolic tangent mean velocity distribution, upon which low amplitude perturbations are superimposed in order to promote the formation of coherent vortices. The initialization of the Shvab–Zeldovich variable at the inflow is such that it takes on the values of zero and one in the streams containing B and A, respectively. The disturbances on the mean flow correspond to the most unstable mode of the hyperbolic tangent profile (Michalke, 1965) and its first two subharmonics. The amplitude of the fluctuating velocity is set equal to 6% of the mean velocity. This amplitude corresponds to that measured in typical shear layer experiments. The magnitudes of the phase shifts between the modes of the instability waves are randomly selected from a random seed with a top hat PDF of zero mean and a specified variance. The implementation of these phase shifts is the only mechanism to introduce randomness into an otherwise deterministic simulation. This is to partially mimic the random "commotions" which are present in the "universe," into an isolated two-dimensional simulation. At the outflow, a weak condition of zero second derivative is applied for all the dependent variables. This boundary condition cannot be justified in a rigorous mathematical sense and can be specified only in an *ad hoc* manner. This was done in order to facilitate implementation of the Dirichlet boundary conditions in the finite element procedure employed here. Moreover, the results of extensive numerical tests showed that the effects of the approximate boundary conditions are confined within the last computational element. This is consistent with that found previously by Korczak and Hu (1987). Therefore, the solution in the last computational domain can be ignored.

With the solution of the unsteady transport equations, the DNS data are extremely useful in visualizing the instantaneous behavior of the flow. The results of these direct simulations can also provide an assessment of the statistical behavior of the flow. This is due to the availability of flow information at all the computational grid points and at all times. Therefore, statistical sampling can be done quite easily by ensemble averaging of the instantaneous data gathered over many realizations.

In our simulations the instantaneous data of the Shvab–Zeldovich variable, obtained from the DNS, are used for statistical analysis. A total of 3200 realizations were employed in the sampling. With this ensemble, the magnitudes of the mean and the variance of the Shvab–Zeldovich variable are calculated at all the grid points. These statistical quantities are of primary interest for turbulence modeling. Based on the knowledge of these first two moments, a Beta density is assumed for the PDF of the Shvab–Zeldovich variable. The ensemble mean values of the product concentration and the unmixedness are calculated subsequently via this density. These modeled quantities are then compared with those obtained directly from the DNS data. At the majority of grid points the Beta density is asymmetric. Nevertheless, analytical solutions for the statistics of the reacting field are possible and have recently been obtained by Madnia *et al.* (1991b). The derivations of the equations are rather involved. Here, we only

present the final results for the normalized mean product concentration $\langle C_p \rangle$ and that of the unmixedness Ψ^2:

$$\langle C_p \rangle = \langle C_p \rangle(x, y) = 1 - \theta_1 - \theta_2 \tag{1}$$

$$\Psi^2 = \Psi^2(x, y) = -\theta_1\theta_2 \tag{2}$$

where:

$$\theta_1 = \frac{1}{2^{\alpha+\beta-1}(\alpha+\beta)} \frac{\Gamma(\alpha+\beta)}{\Gamma(\alpha)\Gamma(\beta)} + \frac{\alpha-\beta}{\alpha+\beta}(1 - I_{\mathcal{I}_n}(\alpha, \beta)) \tag{3}$$

$$\theta_2 = \frac{1}{2^{\alpha+\beta-1}(\alpha+\beta)} \frac{\Gamma(\alpha+\beta)}{\Gamma(\alpha)\Gamma(\beta)} + \frac{\beta-\alpha}{\alpha+\beta} I_{\mathcal{I}_n}(\alpha, \beta) \tag{4}$$

Here, Γ is the Gamma function, and α and β are related to the first two moments of the random field (Frankel *et al.*, 1991). $I_{\mathcal{I}_n}(\alpha, \beta)$ is the Incomplete Beta Function, and $\mathcal{I}_{st}$ denotes the stoichiometric value of the Shvab–Zeldovich variable, $\mathcal{I}$. For unity normalized concentrations at the free streams, $\mathcal{I}_{st} = 0.5$. The results predicted by these equations shall be compared with those generated by DNS for a quantitative assessment of the closure.

PRESENTATATION OF RESULTS

Computations were performed in a domain of size $13\delta \times 34\delta$, where δ denotes the vorticity thickness at the inlet of the flow. There are 65 Fourier modes in the cross-stream direction and 42 finite elements are used for the streamwise discretization. A fifth order Tchebysheff polynomial is employed to approximate the variables within each element. This discretization is equivalent to, at least, a fifth order accurate finite difference technique even if the spectral convergence is not considered. This results in a total number of 211 points in the streamwise direction. A second-order Adams-Bashforth finite difference scheme was utilized for temporal discretization. With the computationally affordable 211×65 grid resolution available, accurate simulations with $Re = Pe = 200$ (where these normalized quantities are defined based on the velocity difference across the layer, and the initial vorticity thickness) are possible (Givi, 1989). These values are somewhat smaller than those of a fully developed laboratory turbulent shear layer, but are sufficiently high to promote the growth of instability waves.

In order to visualize the flow evolution, a time series of DNS generated snapshots of the instantaneously normalized product concentration are shown in Figure 1. This figure depicts the way in which the small amplitude perturbations manifest themselves in the formation of large scale coherent vortices downstream of the splitter plate. The forcing associated with the most unstable mode alone would cause the initial roll-up of these vortices. The perturbations corresponding to the first subharmonic mode result in a second roll-up in the form of merging neighboring vortices, and the presence of the second subharmonic generates a
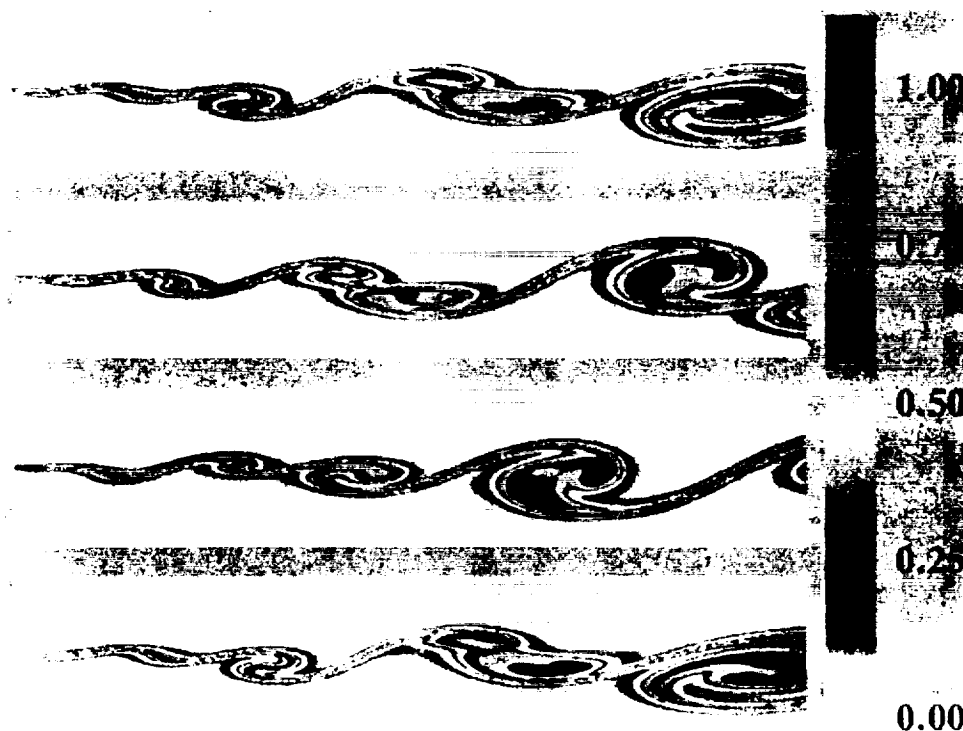
FIGURE 1   Time sequence of contour plots of instantaneous product concentration.
(See color Plate I.)

third roll-up (second merging) at a region near the outflow. The roll-up and the subsequent pairings of these vortical structures engulf the unreacted species from the two streams into the mixing zone. The dynamics of the vortical evolution in this process reflect the two main mechanisms of mixing enhancement. Large concentrations of vorticity, that are approximately of one sign, bring the fluid from the two free streams into the large scale structures. Within these structures, the fluid elements are subject to further straining, and diffusion between the two fluids results in final mixing at the molecular scale. With these processes, along with the assumption of infinitely fast chemistry, the rate of product formation is naturally enhanced by the dynamics of vortical evolution.

The ensemble average of the product concentration, formed as a result of the mechanisms just described, is presented in Figure 2(a). The statistical averaging process was performed over a time span approximately equal to the residence time of the flow within the domain of computational consideration. This analysis was performed after the effects of the initial transients were washed away at the outflow. A comparison between Figures 1 and 2(a) reveals the essential deficiencies of the ensemble averaged results. Note that, all the detailed intricacies of the dynamics of flow evolution are masked by the averaging. While these ensemble-averaged results are of main interest for practical applications,
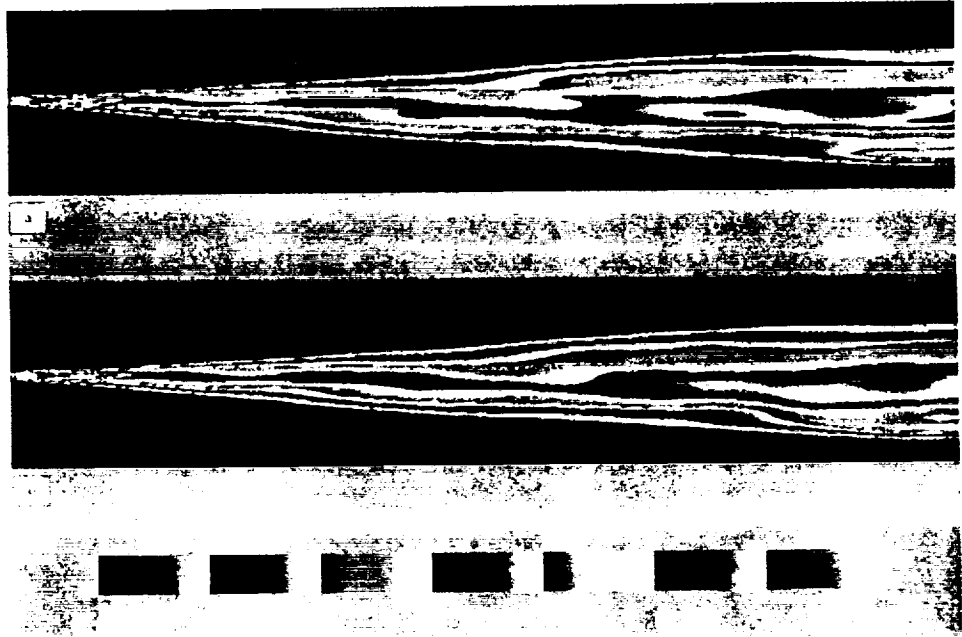
FIGURE 2   Contour plots of mean product concentration generated by (a) DNS, (b) the model.
(See color Plate II.)

they are not capable of describing many of the interesting features of turbulence transport. The results depicted in Figure 2(a) are the best that one can expect to generate from a turbulence closure in a statistical sense.

To demonstrate the capability of the model based on the Beta density, in Figure 2(b), the average product concentration obtained by means of Eqs. (1)–(4) is presented. A comparison of this figure with Figure 2(a) reveals a remarkable similarity between the two results. For a more quantitative comparison, the cross stream variation of this product concentration and those of the negative of the unmixedness are presented at two streamwise locations in Figures 3–4. Also the streamwise variation of the total product, T_p, is presented in Figure 5. In all these figures, the comparisons between the model predictions and the DNS results are noteworthy.

The agreements demonstrated by the comparisons noted above provide a reasonable justification for recommending Eqs. (1)–(4) as working relations in routine engineering predictions. However, these relations are advocated only for statistical predictions of low order moments. The physics of the mixing phenomenon in a spatially developing shear flow is far too complex to be *completely* described by the Beta density. The highly intermittent nature of the mixing process exhibits features that cannot be fully captured by this density (Givi, 1989). To demonstrate this point quantitatively, in Figures 6–7 the cross stream variations of the third and fourth centralized moments of the Shvab Zeldovich variable calculated from the Beta density are compared with those generated by

(a)

(b)
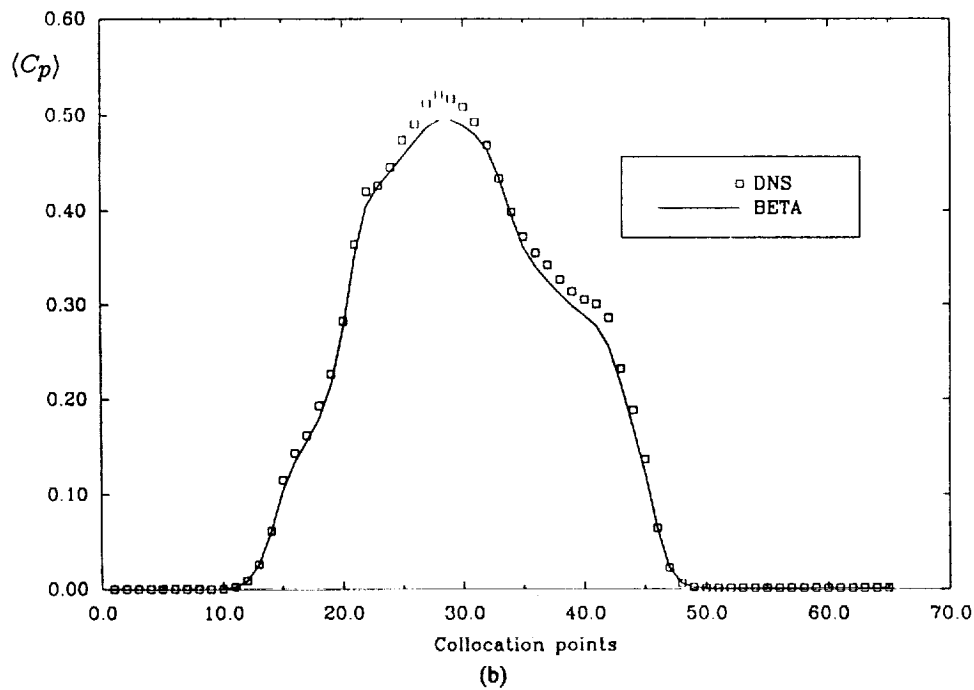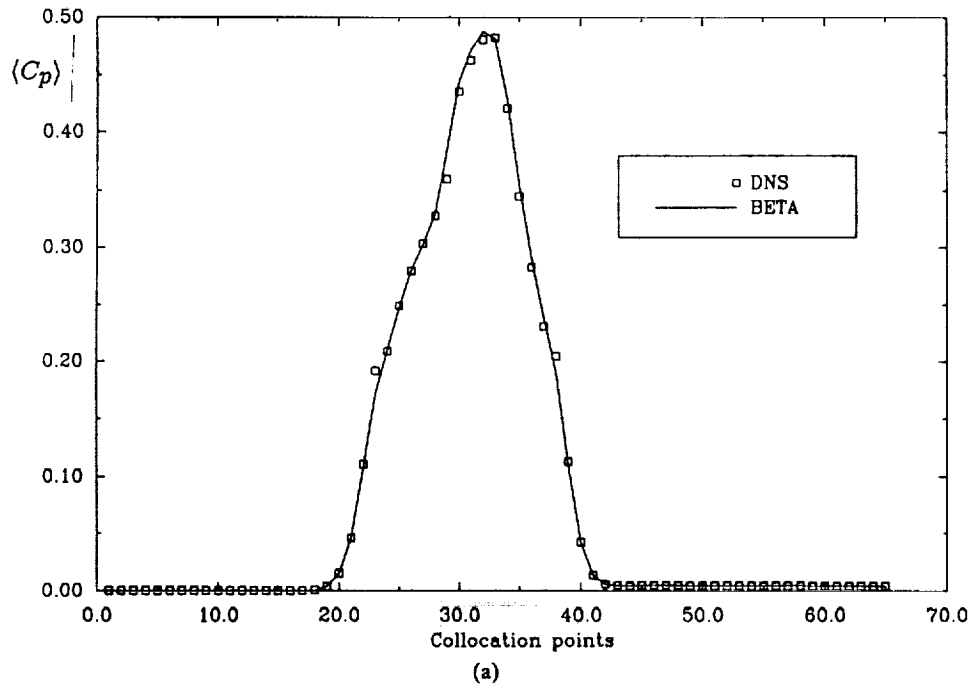
FIGURE 3 Cross stream variation of mean product concentration ($\langle C_p \rangle$) from the DNS and the model; (a) $x/\delta = 11$, (b) $x/\delta - 22$.
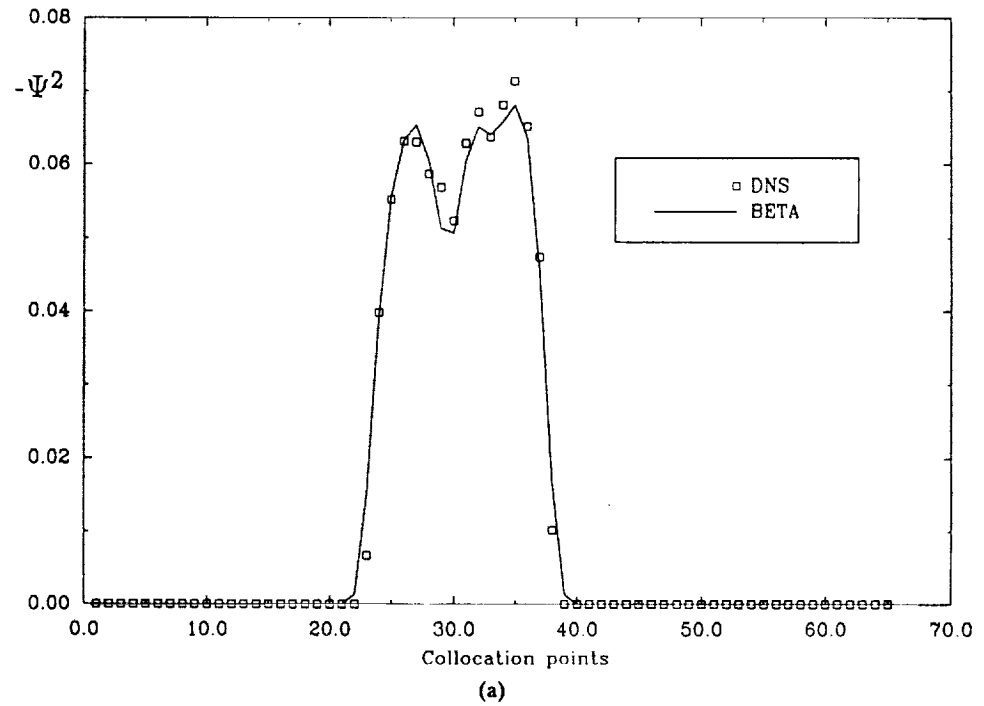
(a)

(b)
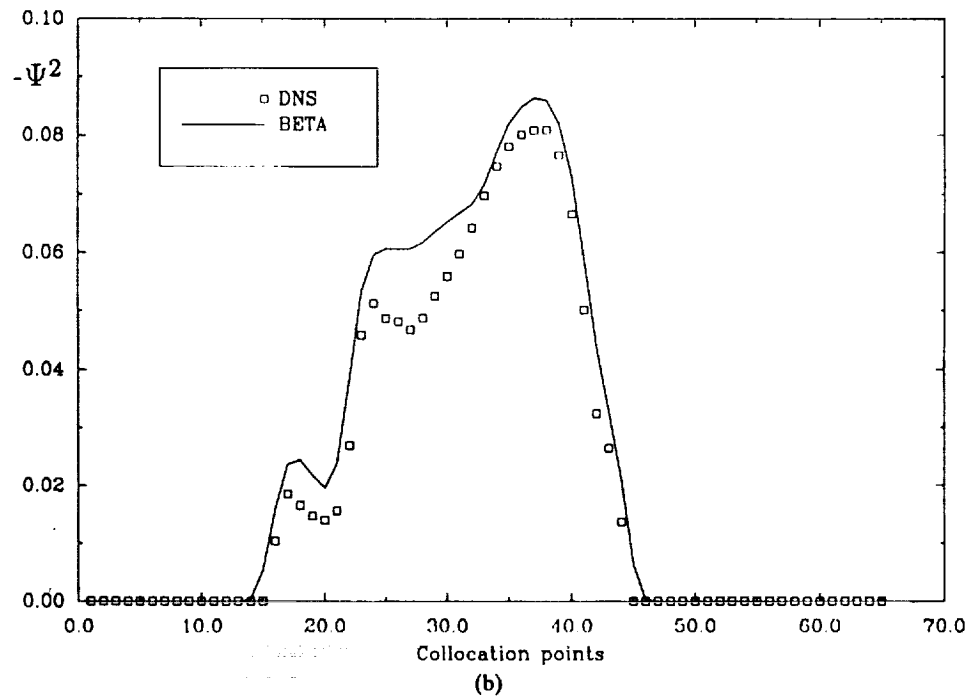
FIGURE 4   Cross stream variation of the negative of the unmixedness $(-\Psi^2)$ from the DNS and the model; (a) $x/\delta = 11$, (b) $x/\delta = 22$.
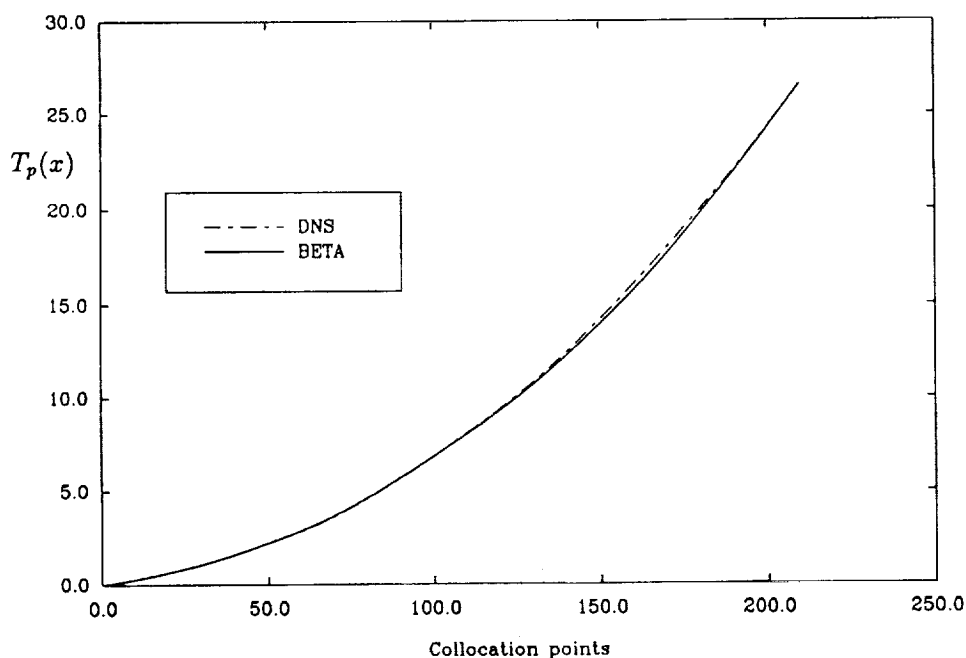
FIGURE 5   Streamwise variation of total product (T_p) from the DNS and the model.

DNS. These figures indicate that while the trend is somewhat similar in the two cases, the extent of agreement is not as good as those of lower level statistical quantities (Figures 2–5). This is primarily due to the mechanism of large scale entrainment in the shear flow, in that there are always unmixed fluids present within the core of the vortical structures. This results in a slight trimodal behavior in the probability distributions generated by DNS (Givi and Jou, 1988; Lowery and Reynolds, 1990) and observed experimentally (Koochesfahani, 1984, Dimotakis, 1989). This trimodal behavior cannot be captured by the Beta density which is at best suitable for producing bimodal distributions.

Recall that the specification of the Beta density is based on the knowledge of the first two moments of the conserved scalar. Here, these moments were supplied via DNS, but in an actual engineering application they must be provided by other means (i.e. an appropriate turbulence closure, or experimental data). However, specification of these parameters is substantially simpler than those of the reacting scalars, even with the assumption of an infinitely fast reaction. This is simply due to the fact that such data can be obtained in simpler, non-reacting flow experiments without any physical complications associated with the chemistry. With such data available, then Eqs. (1)–(4) are deemed very suitable, at least in the absence of better alternatives, for a reasonably accurate and inexpensive prediction of the limiting bounds of the reactant conversion rate. These equations are therefore recommended for this purpose in either homogeneous or non-homogeneous flows.
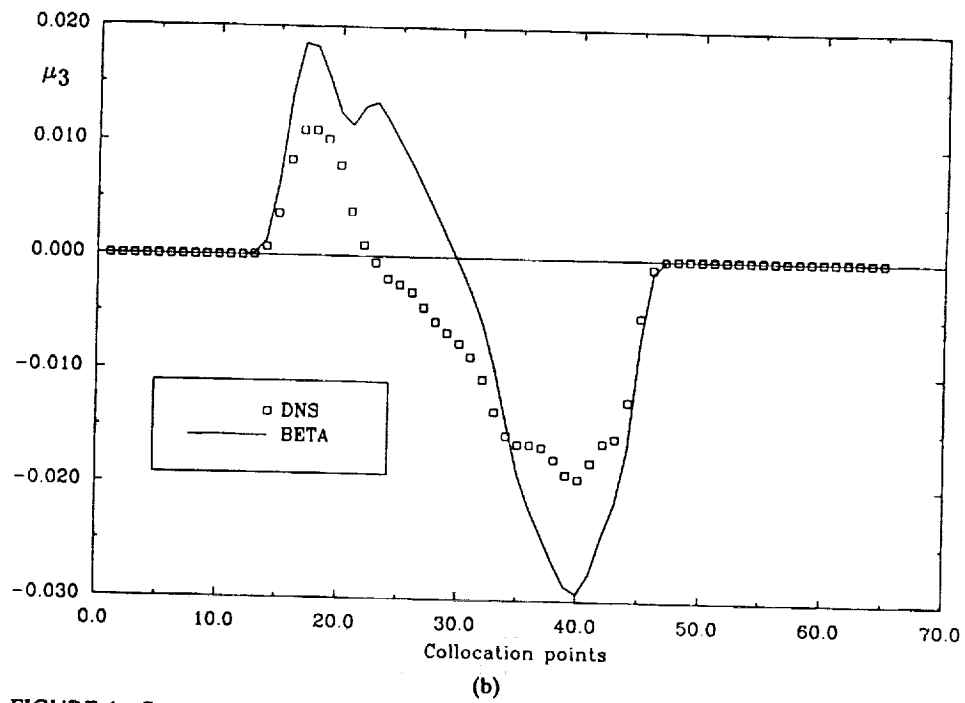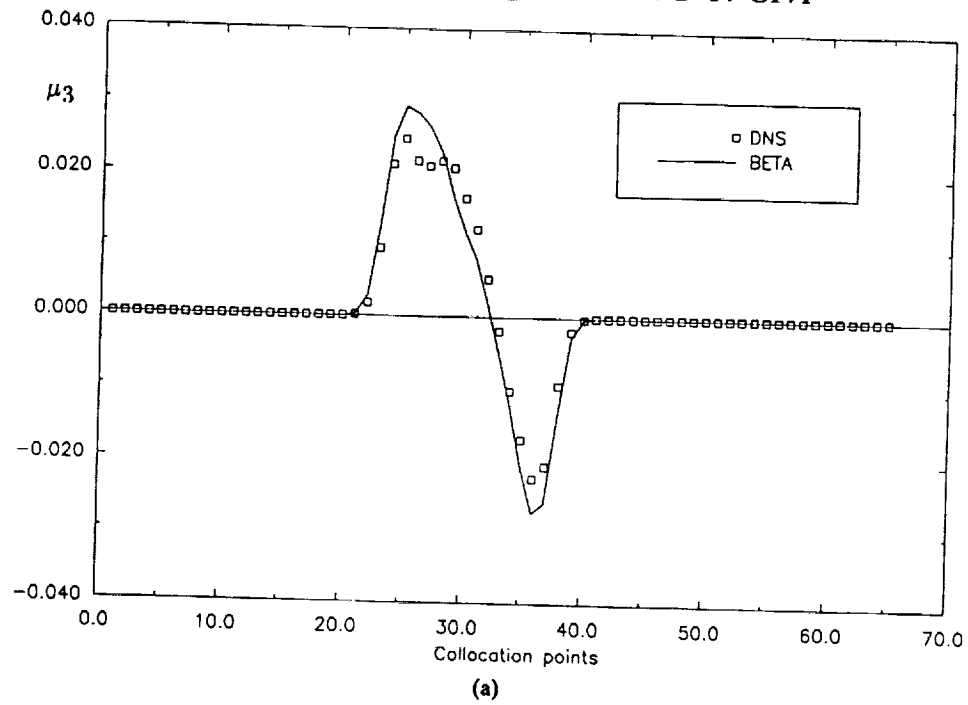
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(a)

(b)

FIGURE 6 Cross stream variations of the third moment (μ_3) of the Shvab–Zeldovich variable from the DNS and the model; (a) $x/\delta = 11$. (b) $x/\delta = 22$.
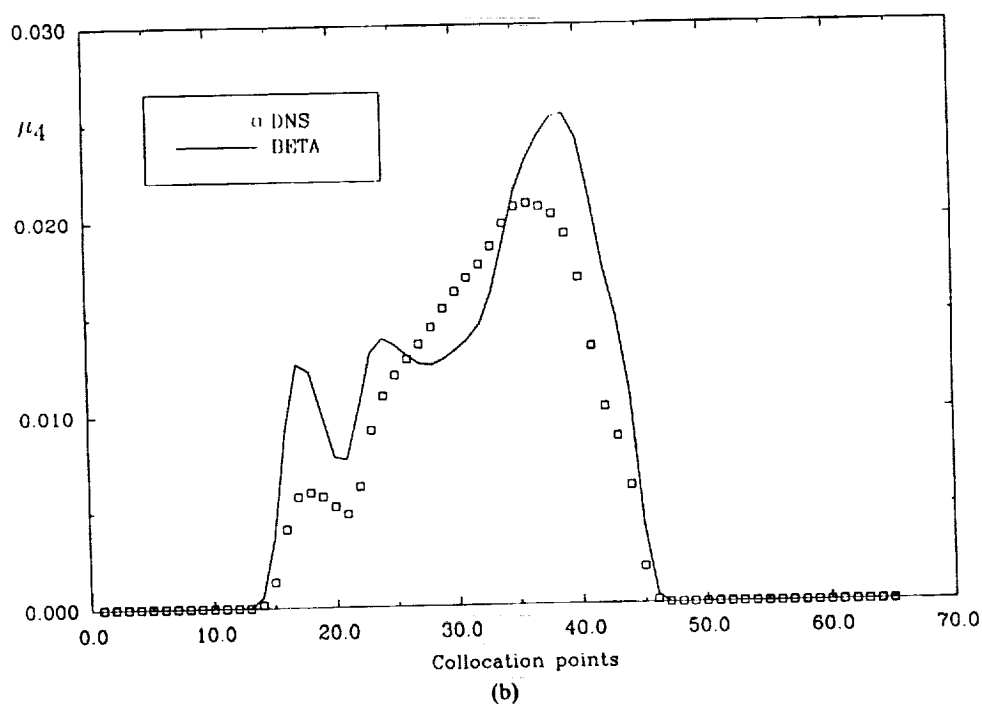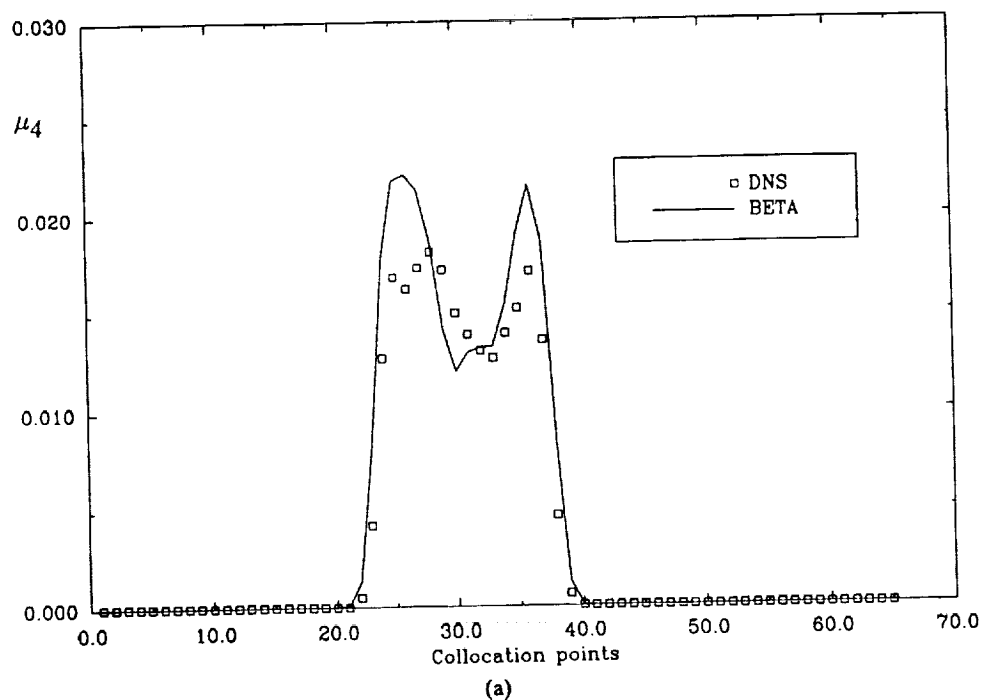
FIGURE 7 Cross stream variations of fourth moment (μ_4) of the Shvab–Zeldovich variable from the DNS and the model; (a) $x/\delta = 11$. (b) $x/\delta = 22$.

ACKNOWLEDGEMENTS

NOMENCLATURE

A,B	Reactants.
C_p	Product Concentration.
$\mathscr{J}$	Shvab–Zeldovich variable.
Pe	The Peclet number.
Re	The Reynolds number.
$T_p(x)$	Total product $= \int_{-\infty}^{+\infty} dy \int_0^x dx \langle C_p \rangle (x, y)$.
x,y	The physical coordinates.
δ	Vorticity thickness at the inflow.
Ψ^2	The unmixedness.
μ_3	Third order moments of the Shvab–Zeldovich variable.
μ_4	Fourth order moments of the Shvab–Zeldovich variable.
$\langle \rangle$	Ensemble average.

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The Compositional Structure and the Effects of Exothermicity in an Unpremixed Planar Jet Flame

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Abstract

Results are presented of direct numerical simulations (DNS) of a randomly perturbed compressible, spatially developing planar jet under the influence of a finite rate Arrhenius chemical reaction of the type $F + O \rightarrow$ Product + Heat. The objectives of the simulations are to assess the compositional structure of the flame, and to determine the influence of reaction exothermicity by means of statistical sampling of the data generated by DNS. It is shown that even with this idealized kinetics model, the simulated results exhibit features in accord with experimental data. These results indicate that the Damköhler number is an important parameter in determining the statistical composition of the reacting field, and that the results are insensitive to the mechanism by which this parameter is varied. It is demonstrated that as the intensity of mixing is increased and the effect of finite rate chemistry is more pronounced, the magnitudes of the ensemble mean and rms of the product mass fraction decrease, and those of the reactants mass fraction increase. Also, at higher mixing rates the joint probability density functions of the reactants' mass fractions shift towards higher values within the composition domain indicating a lower reactedness. These trends are consistent with those observed experimentally and are very useful in portraying the statistical structure of non-equilibrium diffusion flames. The DNS generated data are also utilized to examine the applicability of the "laminar diffusion flamelet model" in predicting the rate of the reactant conversion with finite rate chemistry. This examination indicates that the bounds of the product formation scale reasonably well with those obtained by the flamelet model. However, in the range of the relatively low values of the Damköhler numbers considered, the scatter of the results is substantially more than that to be completely determined by the closure. Finally, the simulated results

suggest that in the setting of a "turbulent" flame, the effect of the heat liberated by
the chemical reaction is to *increase* the rate of reactant conversion. This finding is
different from that of earlier DNS results and laboratory investigations which indicate
a suppressed chemical reaction with increasing heat release.

1 Introduction

Recent advances in flow visualization and diagnostic techniques have made significant con-

tributions to the understanding of turbulent combustion phenomena [1, 2]. With implemen-

tation of these advanced techniques it is now possible to make a detailed examination of

the structure of turbulent flames, and to establish a better understanding of the intricate

physics of such flames. Amongst recent applications of these diagnostic techniques, Masri *et*

al. [3, 4] have made noteworthy progress in describing the structure of unpremixed turbulent

jet flames under the influence of finite rate chemical reactions (see [5, 6] for comprehensive

reviews). In particular, they have reported the results provided by statistical sampling of

the data obtained by Raman-Rayleigh scattering measurements of the species mass fraction

and the temperature in an unpremixed turbulent methane jet flame. These measurements

are utilized primarily for constructing single-point statistics of the reacting field, includ-

ing the ensemble mean, rms and the probability density functions (PDF's) of the relevant

thermochemical variables. The response of the flow under different mixing conditions is

characterized by a comparison of the results obtained at various mixing rates. The sampling

of the data in this way allows a systematic assessment of the compositional structure of the

non-equilibrium flame as it approaches extinction. Such data have proven very valuable in

describing the phenomena of mixing and chemical reactions in unpremixed turbulent combustion, and in addressing some of the existing problems in the modeling of such flames by means of statistical methods.

In order to provide a computational complement to laboratory investigations, there has been significant progress in the development and implementation of direct numerical simulations (DNS) of unpremixed reacting flows [7, 8, 9, 10, 11, 12, 13, 14] (see [15, 16, 17, 18, 19] for recent reviews). The extent of progress made by these contributions has been very encouraging, thus justifying further utilization of direct methods in the analysis of turbulent flames. In this work, we intend to continue our investigation of turbulent flames via DNS, and to simulate such flames in a context relevant to laboratory investigations. Obviously, it is still impossible to perform DNS of a realistic "physical" flame [15]. Here, our intention is to make use of the present capabilities of supercomputers to further address some of the fundamental and pertinent issues in regard to the dynamics of such flames.

In the next section, the method of investigation is presented together with the underlying assumptions imposed in making the problem tractable for computational treatment. The ramifications associated with the simplifying assumptions are discussed, where appropriate, in the presentation of results. This presentation is made in Section 3 with a focus on assessing the compositional structure of the flame, and on revealing the influences of reaction exothermicity on the evolution of the flame. With this presentation, it is shown that some of the trends observed in laboratory experiments can be captured by DNS, and speculation is made on some of the other features not yet considered in laboratory investigations. This paper is drawn to a conclusion in Section 4.

2 Method of Investigation

The flow under investigation consists of a spatially developing, two-dimensional, planar jet flame. The schematic diagram of the jet is shown in Fig. 1(a) together with the specifications of the physical dimensions. The fuel, F, discharges from the inner higher speed jet of width D with a velocity U_F into a co-flowing lower velocity (U_O) oxidizer, O, stream. The inlet temperature and pressure are the same at the inlet of both streams, and the reactants are completely segregated at the inlet. This configuration is different from that of a circular jet flame considered in [3, 4], but allows the elucidation of some specific physical features that are believed to be invariant of the jet geometry [18]. The flow evolves spatially in the streamwise direction (x), and impermeable boundary conditions are imposed in the cross-stream direction (y). The DNS computational procedure is the same as that employed in [14, 20]. This involves the discretization of the complete Navier-Stokes equations as well as the applicable Fickian chemical species conservation equations. This discretization is based on a *two-four* (second order accurate in time, and fourth-order truncation in space) compact parameter finite difference scheme. This scheme has been developed and made fully operational by Carpenter [21] and is utilized in the context of the *SPARK* computer code developed by Drummond [22]. A generalized single step Arrhenius exothermic chemistry model of the form $F + O \rightarrow$ Product + Heat is assumed. This kinetic mechanism is simple enough to allow an efficient treatment via DNS, and yet it is capable of capturing some of the important non-equilibrium effects associated with combustion. All the species involved in this reaction are assumed to be thermodynamically identical, and the values of the physical transport parameters are assumed to be invariant with temperature. With this kinetics

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prototype, the rate of reactant conversion is expressed by:

$$\omega = K_f C_F C_O \exp(-\frac{E}{\mathcal{R}T})$$ (1)

The flow field is initialized with a top hat streamwise velocity profile with a low amplitude random forcing at the jet entrance. The magnitudes of the forcing frequency and the phase shifts between the different components of the forcing eigenfunctions are selected randomly from a seed with a uniform probability distribution and with specified values of the mean and variance. This random specification of the frequency and the phase shift is intended to mimic the influence of turbulence in an otherwise deterministic simulation. The amplitudes of the perturbation are chosen so that the maximum value of the turbulent intensity at the inflow is no more than 5%.

The parameters influencing the rate of reactant conversion in this setting are the Damköhler number, the Zeldovich number, the Reynolds number, the heat release parameter, the convective Mach number, the velocity ratio between the streams, and the Prandtl/Schmidt numbers (see the nomenclature for a description of the thermochemical variables and for a definition of all the relevant non-dimensionalized parameters). Based on our earlier findings [20, 14], in the range of parameters considered the Reynolds number does not have a significant effect. Therefore, the influences of this parameter as well as those of the Peclet and the Schmidt numbers are not investigated. The values of the molecular Prandtl and the Schmidt numbers are set to unity ($Pr = Sc = 1$), to make the analysis associated with laminar diffusion flamelet modeling easier. The magnitude of the Zeldovich number

is held constant in all the simulations, but the values of the Damköhler numbers and the heat release parameter are altered to assess their influence on the dynamics of combustion. The full compressible form of the transport equations are considered in the computational formulation. However, the magnitudes of the convective Mach numbers considered are not very large. Therefore, many of the physical issues associated with high speed combustion [22, 18, 14] are not addressed. Also, the body forces are assumed negligible; therefore, the interesting physical problems associated with the buoyancy [23, 24] are not considered.

The computer code used is ideal for high resolution, three-dimensional (3-D) simulations in a reasonably efficient manner on presently available supercomputers [16, 20]. However, the specific physical issues considered here do not require 3-D treatment, and can be addressed in a 2-D context. This was established by some comparative assessments of 2-D and 3-D results, and all subsequent simulations were performed in a 2-D domain. This yields a higher numerical resolution at a lower computational cost.

In accordance with the procedure implemented in [3], three flames are considered; Flames A, B, and C. These flames are identified by the relative magnitude of the mixing intensity quantified by the value of the Damköhler number. In Flames B and C, the magnitudes of the Damköhler number are the same and are set equal to half of that in Flame A. However, the mechanism by which this is enacted is different. In Flame B, this is implemented by decreasing the magnitude of the chemical reaction frequency, and in Flame C by increasing the magnitude of the characteristic velocity of the jet. Both of these procedures result in the same reduction in the value of the Damköhler number, and the difference between Flames A and C is the same as that in the flames considered in [3]. The magnitude of the convective

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Mach number is higher in Flame C than that in Flame A. But, the value of this Mach number is not so large as to have a substantial influence on the spatial growth of the jet [14]. Also, the imposition of external forcing masks the effects of other possible mixing inhibition mechanisms (such as exothermicity, density stratification, *etc.*) [12, 24, 14].

The compositional structure of the flame is assessed by means of examining the response of the flow to the Damköhler number. This assessment is quantified by considering the statistical properties of the reacting field, including the ensemble mean, rms and the PDF's of the mass fractions of all the species. The analysis has been made first in a non-heat releasing reacting jet. The important influences of the exothermicity are subsequently considered.

3 Presentation of Results

With available computational resources, numerical experiments with a resolution of 245 (in x) $\times 165$ (in y) mesh are possible. This resolution is sufficient provided that an appropriate grid generation procedure with a non-uniform mesh and with a heavy concentration of grid points near the regions of maximum instantaneous shear is utilized (Fig. 1(b)) [22, 20, 14]. With this configuration, it is possible to perform simulations with moderate values of the Reynolds, Peclet, Damköhler and Zeldovich numbers within a domain $L_x = 13\frac{1}{3}D > x > 0$, $L_y = 6\frac{2}{3}D > y > 0$. With the numerical resolution attained, it was possible to perform simulations with a Reynolds number of $Re \approx 6 \times 10^4$. The magnitude of the Damköhler number is $Da = 5$ for Flame A and $Da = 2.5$ for Flames B and C. The Zeldovich number is

set at $Ze = 20$ for all the flames, and the heat release parameter is in the range $Q = 0 \sim 5$. The velocity ratio between the two streams is $\frac{U_O}{U_F} = \frac{1}{2}$. With the resolution adopted, a typical simulation requires about 6 hours of CPU time on a CRAY-2 supercomputer.

To visualize the global structures and the unsteady evolution of the layer, a time sequence of the DNS generated instantaneous product mass fractions is shown in Fig. 2(a). This figure shows how the growth of low amplitude random perturbations manifest themselves in the formation of large scale coherent vortices downstream of the jet entrance. Due to the random nature of the forcing mechanism, the evolution of the large scale structures is very complex and, similar to laboratory shear flows, consists of random formation of vorticity rollups and subsequent pairing and coalescence of the neighboring vortices. Also, due to this randomness, the layer is asymmetric with respect to the jet centerline even though a symmetric disturbance is imposed at the inflow. In all the simulations, the energy spectrum of the fluctuating hydrodynamics field is fairly broad-banded. Therefore, the flow can be assumed "turbulent", albeit two-dimensional. The formation of large scale structures results in an enhancement of mixing, as demonstrated in Fig. 2(a). The unsteady conformation of large scale structures results in the mutual engulfment and entrainment of the two reactants, and the dynamics of this evolution reflects the primary mechanisms of the mixing process. Large concentrations of vorticity on both sides of the jet centerline, bring the fluid from the two jets into the core of the large scale structures. Within these structures, the fluid elements are subject to further straining, and the molecular diffusion between the two fluids results in final mixing at the molecular scale. The enhanced mixing mechanism caused by the large scale vortices results in a general augmentation of the product formation. At

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regions close to the jet entrance, the chemical reaction is promoted by the formation of coherent vortices, and there is a substantial amount of product formed. If the flow was in chemical equilibrium, the total amount of product would have increased monotonically along the streamwise direction. For the non-equilibrium chemistry considered here, this is not the case and mixing enhancement does not necessarily yield improved combustion. It is shown in Fig. 2(a) that at the shear zone near the entrance of the jet, where the reactants are first brought into contact, the extent of chemical reaction is fairly uniform. Further downstream, as the magnitude of the instantaneous strain increases, the reaction rate approaches zero at the braids of the vortices and the flame locally quenches at those locations. This mechanism of flame extinction is consistent with previous DNS results [9, 10], and is also corroborated by experimental observations [25, 26]. This behavior was observed in all the simulations with moderate Damköhler numbers asserting the departure from chemical equilibrium.

The effects of the non-equilibrium chemistry become more clear as the influence of finite rate kinetics is more pronounced. To show this influence in the format adopted in [3], the results extracted by DNS are statistically analyzed. With the assumption of statistically stationary flow, the instantaneous data can be used to construct ensemble-averages. Here, these averages are obtained by statistical analysis of 1250 time samples gathered after the effects of initial transients are washed away at the outflow. The data sampling was performed within a time period equivalent to half the residence time of the flow, which was shown to yield statistically acceptable trends. These results are presented in Fig. 2(b) in the form of contour plots of ensemble-averaged product mass fraction. A comparison between the two parts of Fig. 2 reveals the essential deficiencies of the ensemble averaged results. Note

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that the intricate dynamics of the flow evolution are masked by the averaging procedure. While these ensemble-averaged results are of interest for practical applications, they are not capable of describing the interesting detailed features of turbulent transport. Nevertheless, the results presented in Fig. 2(b) are those which one can expect to generate by using a turbulence closure. Therefore, an assessment of their validity is essential for the purpose of statistical modeling. These results also facilitate the most convenient (and perhaps the most traditional) means of comparing our DNS data with experimental measurements.

To exhibit the effects of finite rate chemistry in a quantitative manner, the statistical analysis of the data is very useful. With DNS data available at every time step at all the locations within the flowfield, an extensive library of data sets is produced. In the discussions below, the statistical data are presented at several streamwise locations where the appropriate physics are captured. In Fig. 3 the cross stream variation of the ensemble-mean values of the product mass fraction is presented at a streamwise location ($x = 10D$). This figure shows that the magnitude of mean product mass fraction in Flame A is substantially higher than those of Flames B and C. This suggests that as the flame approaches extinction, the mass fractions of the reactants in the reaction zone increase and, as a result of local flame quenching, the rate of product formation decreases. The same behavior is observed in Fig. 4, which shows that the amplitude of the rms of the product mass fraction is higher in Flame A than those in Flames B and C. The trends observed in these two figures are consistent with those of the laboratory measurements [3] and reveal the effects of the mixing intensity on compositional structure of the flame. These results also indicate that this behavior is observed regardless of the procedure by which the Damköhler number is varied. In the experiments,

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the Damköhler number was altered only by means of increasing the hydrodynamic intensity (*i.e.* increasing the inner jet velocity). This was because a single fuel was used and, therefore, the chemical frequency was constant in all the experiments. Our results indicate that this dependence is observed regardless of the procedure by which the change is made, and Flames B and C portray approximately the same statistical conduct. The behavior observed in Figs. 5-6 demonstrates the dominant influence of the Damköhler number in quantitative analysis of non-equilibrium flames, and also in the comparative assessment of the statistical results generated in our DNS with those obtained in laboratory experiments [3] (See [27] for more discussions pertinent to this issue).

The influence of finite rate chemical reactions on the compositional structure of the flame is further demonstrated by examining the marginal and the joint PDF's of the reacting scalar variables, as well as the "scatter" plots of the product mass fraction. These are presented in Figs. 5-8. In Fig. 5, results are presented of typical joint PDF's of the Fuel and the Oxidizer mass fractions, *i.e.* $\mathcal{P}(Y_F, Y_O)$ for Flames A and B. This figure indicates that at lower mixing rate, Flame A (Fig. 5(a)), there is a larger amount of product formed, and the height of the PDF's are larger at the mixed mass fraction values at the interior of the allowable region ($0 \leq Y_A, Y_B \leq 1$). At the higher mixing rate, Flame B (Fig. 5(b)), the mixture is composed of the two reactants with the joint PDF's concentrated primarily along the line of $Y_F + Y_O = 1$. These results are consistent with those reported in [4] and indicate that at the higher levels of mixing intensity, the results are closer to those obtained by cold mixing.

The corresponding marginal PDF's of the product mass fraction $\mathcal{P}(Y_p)$ are shown in Figs. 6

for Flames A and B. In both flames, the PDF's are concentrated near zero at free streams, and become broader near the central region of the layer. As the mixing rate is decreased, the PDF's are shifted toward higher values of the mass fraction. This behavior is also consistent with the experimental data on PDF's of product species, $H_2, CO_2, \ldots$ [4] and indicate lower reactedness at higher mixing rates. However, due to strong intermittency of the flow in the two-dimensional simulations, the PDF's do not adopt a Gaussian distribution, and are always heavily populated near free stream values, $i.e.$ zero. Also, the tails of the PDF's get thinner as the mixing is intensified. This suggests a reduction in the rms of the product mass fraction, as was reported in the experiments of Masri $et\ al.$ [3], and also evident in Fig. 4.

The lesser rate of product formation is also evident in the scatter plots of the product mass fraction vs. the mixture fraction in Figs. 7 and 8. These figures represent conditions at several streamwise locations, including all the values in the cross stream direction. There are a total of 102500 data points taken in devising these scatter plots. It is indicated in these figures that the rate of product formation is generally high near the inflow, and gradually decreases further downstream as the influence of flame extinction becomes more pronounced. Note that at $x = 6.8D$, the reaction zone is confined to a small region near the equilibrium limit, but at distances further downstream the flame becomes widely distributed within the allowable region. This spread is more noticeable for the lower Damköher number flame, indicative of the pronounced effects of the extinction. This is to be expected, since in the context of the simplified kinetics model, it is expected to have an infinitely thin reaction zone near the equilibrium limit, as the magnitude of the Damköhler number becomes infinitely large. With a departure from this limiting rate, the scatter becomes wider as the Damköhler

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number is reduced. Also, as the influence of flame extinction becomes more pronounced (at lower Damköhler numbers), it is not surprising to have a lower rate of product formation, as evidenced by the smaller amplitudes of the mass fraction values in Fig. 8 in comparison with those in Fig. 7.

In order to have a quantitative means of departure from the equilibrium, the simulated results in Figs. 7-8 are also compared with those predicted by the steady laminar diffusion flamelet model proposed in [26, 28]. For this comparison, the simplified chemical kinetics model employed above was also considered in the content of a simple one-dimensional opposed jet laminar flame configuration similar to those in [29, 30, 31]. In this setting, with the assumption of unity Lewis number, the simulations based on the flamelet model involves the solution of the diffusion-reaction equation of type [31]:

$$\frac{d^2\psi}{d\xi^2} = \frac{-2\omega_\psi}{\rho\chi}.$$

(2)

In this equation, ψ denotes the reacting scalar variable, *i.e.* species mass fractions, and the temperature, with the corresponding chemical source term, ω_ψ. The variable χ represents the dissipation of the conserved mixture fraction, ξ, is expressed by:

$$\chi = \chi_{st} \exp\left[-2\left(\text{erf}^{-1}(2\xi - 1)\right)^2\right].$$

(3)

Here, χ_{st} denotes the "stagnation" value of the scalar dissipation. In the laminar jet flame,

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this stagnation value is determined directly by the hydrodynamic intensity of the jet. However, for the unsteady, "turbulent" jet flame considered here, a single value cannot be adopted. Instead, only some representative values are considered. The results corresponding to several of these values are superimposed on Fig. 7. Obviously, as the magnitude of the stagnation dissipation increases, the rate of fuel consumption decreases, and the results are always bounded in the upper limit as the value of the Damköhler number approaches infinity. Now, holding the value of the stagnation dissipation constant together with the other important kinetics parameters, but decreasing the value of the Damköhler number, the results can be superimposed on the scatter plots in Fig. 8. With the reduction of the Damköhler number the rate of product formation becomes less, and the laminar diffusion flamelet model shows similar trends as those of the DNS data. However, as shown in both Figs. 7 and 8, with a fixed pre-determined value of the stagnation dissipation it is not possible to capture the complete picture, as the predicted results cannot account for the wide scatter in the figures. The comparison between the model predictions and the DNS results would probably be better at higher Damköhler numbers, with a thinner reaction zone, and perhaps by employing a "variable" form for the stagnation scalar dissipation. It is not presently clear how to modify this rate to account for the strong effects of the spatial inhomogeneities, and it is speculated that the functional form of the stagnation dissipation is strongly dependent on the details of the flow field. Nevertheless, the simulated results with this strong finite rate kinetics effects are consistent with the laboratory data, as similar behavior was also observed in the experimental measurements [4].

Despite the attractive features of our simulations in predicting the trends observed experi-

mentally, it must be emphasized that in the context considered, the factors influencing the stability characteristics of the jet are masked by addition of external forcing. Note that these simulations are of a *transitional* flow, whereas the experimental data are obtained in a fully *turbulent* jet flame. The imposition of explicit forcing was therefore necessary to mimic a turbulent environment so that meaningful comparisons with experimental results could be made. This forcing, however, overcomes the mixing suppression caused by factors such as reaction exothermicity, compressibility, *etc.* [14]. In order to make this point clear, simulations are performed of a heat releasing flame (Flame D). This flame has the same characteristics (*i.e.* identical velocities, Damkölher, and Zeldovich numbers) as Flame A, but it also accounts for the effects of exothermicity (*e.g.* a non-zero enthalpy of combustion). A comparison between the total products generated by this flame and that by Flame A is made in Fig. 9. This figure indicates a higher rate of product formation for the case of exothermic reaction. This observation is not consistent with any of the previous DNS [8, 14] or experimental [32] results. The reason for this discrepancy is due to the effects of the Arrhenius kinetics model in the present forced simulations, and *cannot* be observed in constant rate kinetics simulations and laboratory experiments [8, 14, 32]. The increased level of heat release also delays the onset of extinction, since the heat transfer away from the flame is countered by the heat generated inside. This new observation, therefore, suggests that in the setting of a "turbulent" flame in which the "transitional effects" are not dominant, and where the chemical reaction is describable by an Arrhenius model, the heat release results in a *higher* reactant conversion, even though the rate of mixing may be somewhat less.

Based on our observation, we recommend that the effects of exothermicity be assessed by

means of laboratory measurements. These measurements must involve a reacting system whereby the rate of reaction conversion is temperature dependent (unlike that employed in [32]), and in which the large scale mixing intensity is not significantly affected by the heat release (see [11, 12, 17, 24, 19, 18] for discussions on the effects of heat release in low speed combustion, and [14, 20] for discussions of such effects in high speed combustion). It is important to note that the issue would not be resolved by a mere comparison of the mixing characteristics between a reacting and a non-reacting layer. An appropriate analysis requires the consideration of two reacting systems with the same hydrodynamic characteristics but with different magnitudes of the enthalpy of combustion. The extent of validity of our conclusion under more complex chemistry models can be determined by these experiments.

4 Concluding Remarks

A two-four compact differencing algorithm is employed for direct numerical simulations of a spatially developing planar jet flame under non-equilibrium chemical conditions. With the high numerical accuracy provided by this algorithm, it is possible to perform direct simulations under physically realistic conditions of variable density and exothermic reaction. Here, the kinetic mechanism is assumed to obey the idealized model of the type $F + O \rightarrow$ Product + Heat.

The data produced by DNS are statistically sampled to assess the compositional structure of the flame. The results of this analysis are shown to be consistent with those of laboratory

measurements, even with the assumption of the idealized chemistry model. It is shown that as the intensity of mixing increases, and the flame approaches extinction, the magnitudes of the ensemble mean and rms of the product mass fraction decrease whereas those of the reactants increase. This conclusion is only valid if the physical mechanisms by which the growth of instability waves is suppressed are masked. This procedure was implemented here by means of imposing a low amplitude random forcing at the entrance of the jet.

The marginal and the joint PDF's of the reactants mass fractions, extracted from DNS data, show features in accord with laboratory data. That is, as the relative intensity of mixing is increased, and the value of the Damköhler number is decreased, the PDF's show a lesser probability of product formation. The scatter plots of the instantaneous product formation also show trends in accord with experimental data. These results also scale well with those based on the laminar diffusion flamelet model. However, the strong scatter in the results at the low values of the Damköhler number considered, suggest that to achieve a better agreement requires the use of a "flow dependent" form for the scalar dissipation. It is anticipated that the "correct" functional form of this dissipation is strongly dependent on the flow configuration, and it is not clear what form is appropriate for the setting of the unsteady jet flame considered here.

The results further indicate that in the setting of a "turbulent" flame, the exothermicity results in an enhanced reactant conversion and an increased rate of product formation. This trend is not in accordance with previous DNS and laboratory results. The factors resulting in this discrepancy are: (1) imposition of external forcing, (2) the implementation of an Arrhenius kinetic model. Both of these factors are very important in diffusion flames in

which the flow is fully turbulent and the chemical reaction rate can be assumed to obey the Arrhenius law. Therefore, we propose that in typical hydrocarbon turbulent diffusion flames, the effect of heat release is to increase the rate of product formation even though the rate of mixing may be somewhat reduced.

The results of our current preliminary investigations indicate that our major conclusions remain unchanged for 3-D simulations. However, the effects of small scale turbulence motion are very important in the analysis of the compositional structure with a view on flamelet modeling [26, 5, 6]. With such simulations, it is anticipated that the scatter would become more severe. Also, the extraction of high level statistical data, such as the marginal and joint PDF's are more appropriate in the random field of an unsteady three-dimensional configuration, dominated by small scale structures.

Considering the fact the kinetics mechanism employed in the present simulations is very idealized, the qualitative agreement between the experimental measurements and the simulated data is noteworthy.

Symbols

C. Concentration.

c_p. Specific heat of the mixture at constant pressure.

D. The jet width at the inflow.

Da. The Damköher number: $Da = \frac{K_f C_F}{\Delta U / D}$.

E. Activation energy for the Arrhenius chemical reaction.

F. Fuel.

K_f. Pre-exponential factor of the reaction rate.

O. Oxidizer.

Mc. The convective Mach number.

P. Product.

$\mathcal{P}$. Single-Point Probability Density Function.

Pr. The Prandtl number.

Q. Non-dimensionalized heat release parameter $Q = \frac{-\Delta H^0}{C_p T_F}$

$\mathcal{R}$. Universal gas constant.

Re. The Reynolds number. $Re = \frac{\Delta U \rho D}{\nu}$.

Sc. The Schmidt number.

T. Temperature.

U. Streamwise velocity.

x, y. Streamwise and cross stream directions.

Y. Mass fraction.

Ze. The Zeldovich number, $Ze = \frac{E}{\mathcal{R}T}$.

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Greek Symbols, Subscripts and Other Notations

ν. Kinematic viscosity.

ρ. Density.

$\langle\ \rangle$. Ensemble-averaged.

$-\Delta H^\circ$. Enthalpy of combustion.

ΔU. Velocity difference of the two streams $= U_F - U_O$.

χ. Instantaneous dissipation of the conserved mixture fraction.

ψ. Representation of a reacting scalar variable.

ω. Reaction rate.

ω_ψ. Appropriate reaction rate for the scalar variable ψ.

ξ. The mixture fraction.

T_P. Total product mass fraction. $T_P(x) = \int_\infty^\infty Y_P(x,y)dy$.

st. Stagnation.

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6 Appendix II

This work summarizes the first efforts in validating the use of the Pearson family of frequencies in the modeling of non-homogeneous reacting flows. This work was deemed necessary for the following reasons:

1. Before applying the Pearson density for subgrid modeling in LES, this closure must be validated and assessed for RANS of non-homogeneous flows.

2. Because of the similarity of the PDF's produced by the AMC and the Pearson family in homogeneous flows (Appendix I), it is speculated that the same similarity holds in non-homogeneous flows. However, the implementation of the AMC for non-homogeneous flow is very difficult. Therefore, in this work, we have started with the simple model of the Beta density.

Work is in progress for the extension of this work for the LES of non-homogeneous shear flows.

Mr. Steven Frankel and Dr. Cyrus K. Madnia prepared this Appendix.

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7 Appendix III

In this work, Mr. Craig Steinberger, with the help of his colleague, Mr. Thomas Vidoni, has focused his efforts in further investigations of non-equilibrium compressible reacting shear flows. This particular aspect of his work is concerned with: (1) The effects of finite-rated kinetics in statistical description of non-equilibrium diffusion flames, (2) The extent of validity and applicability of the laminar diffusion flamelet modeling for finite rate chemistry diffusion flames, and (3) Further effects of exothermicity in turbulent diffusion flames.

In addition to the subject matters described above, the data bank provided by these simulations would be very useful for further statistical modeling of turbulent flames in future investigations.

[33] Oran, E. S. and Boris, J. P., editors, *Numerical Approaches to Combustion Modeling, Progress in Astronautics and Aeronautics*, Vol. 135, AIAA Publishing Co., New York, NY, 1991.

Figure Captions

Figure 1. (a) Schematic diagram of the planar jet configuration. (b) the grid.

Figure 2 (a). Plot of product mass fraction contours. (a) Instantaneous data at several times $(t_3 > t_2 > t_1)$. (b) Ensemble-averaged data.

Figure 3. The cross-stream variations of the ensemble-averaged product mass fraction at $x = 10D$.

Figure 4. The cross-stream variations of the rms of the product mass fraction at $x = 10D$.

Figure 5. Joint PDF's of the reactants mass fractions, $\mathcal{P}(Y_F, Y_O)$ at $x = 10D, y = 3.8D$. (a) Flame A. (b) Flame B.

Figure 6. Marginal PDF's of product mass fraction $\mathcal{P}(Y_P)$ at $x = 10D, y = 3.8D$. (a) Flame A, (b) Flame B.

Figure 7. Scatter plots of product mass fraction vs. the mixture fractions for Flame A, and comparison with the Laminar Diffusion Flamelet Model. (a) $x = 6.8D$, (b) $x = 7.9D$, (c) $x = 10D$, (d) $x = 11.4D$.

Figure 8. Scatter plots of product mass fraction vs. the mixture fractions for Flame B, and comparison with the Laminar Diffusion Flamelet Model. (a) $x = 6.8D$, (b) $x = 7.9D$, (c) $x = 10D$, (d) $x = 11.4D$.

Figure 9. The streamwise variation of the total product mass fraction $T_P(x)$ under both non-heat releasing (Flame A) and heat releasing (Flame D) conditions.
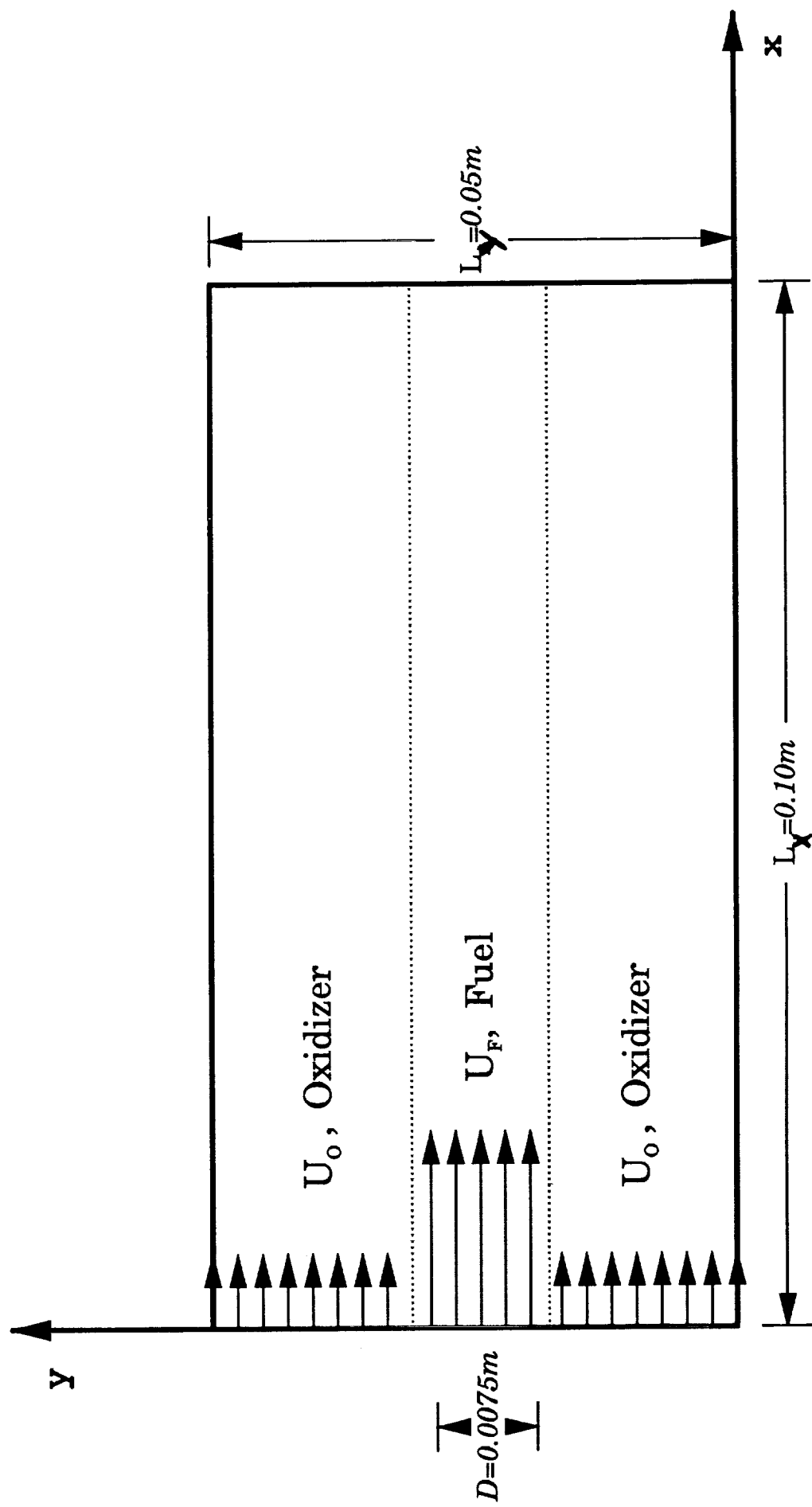
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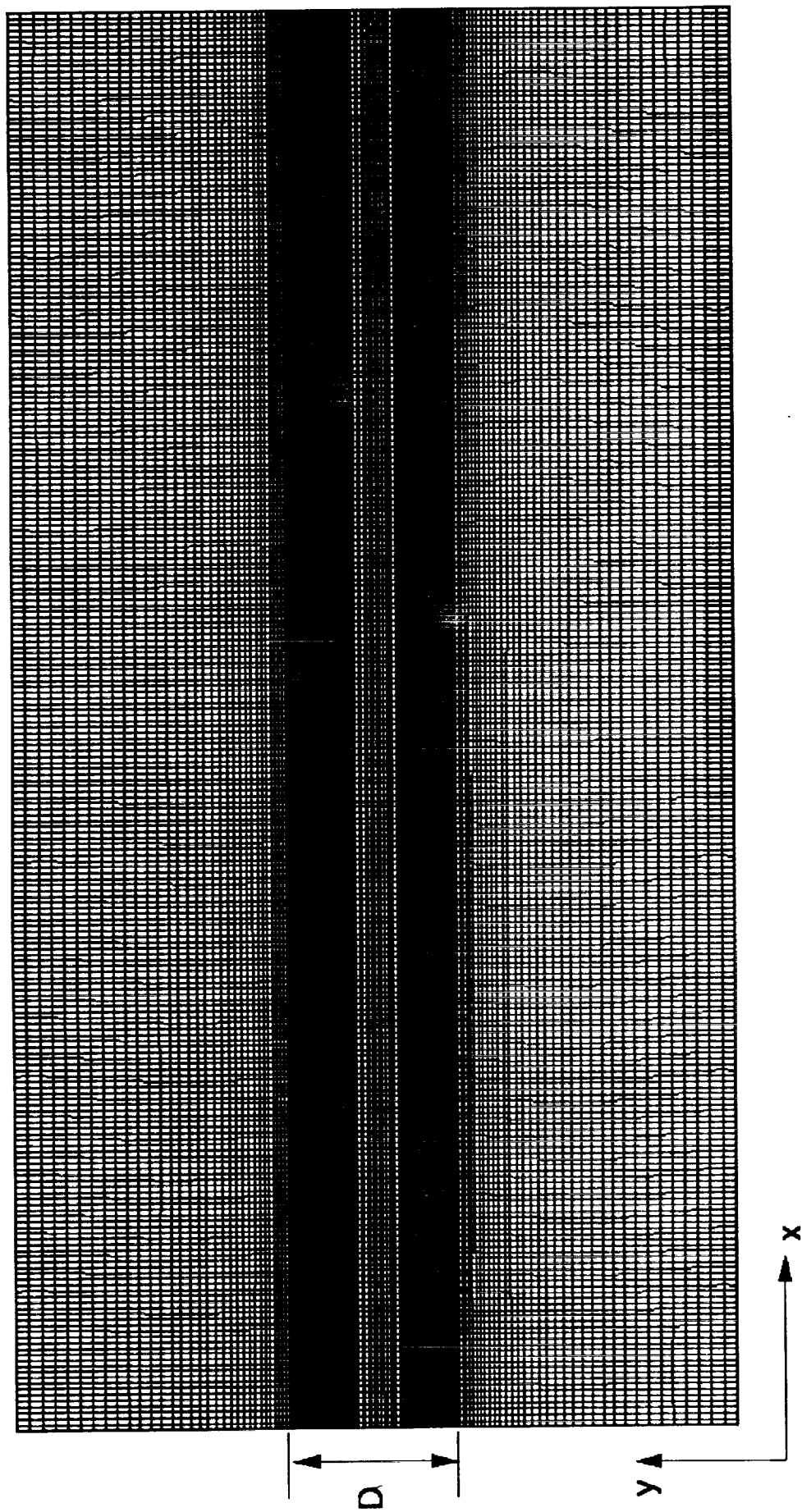
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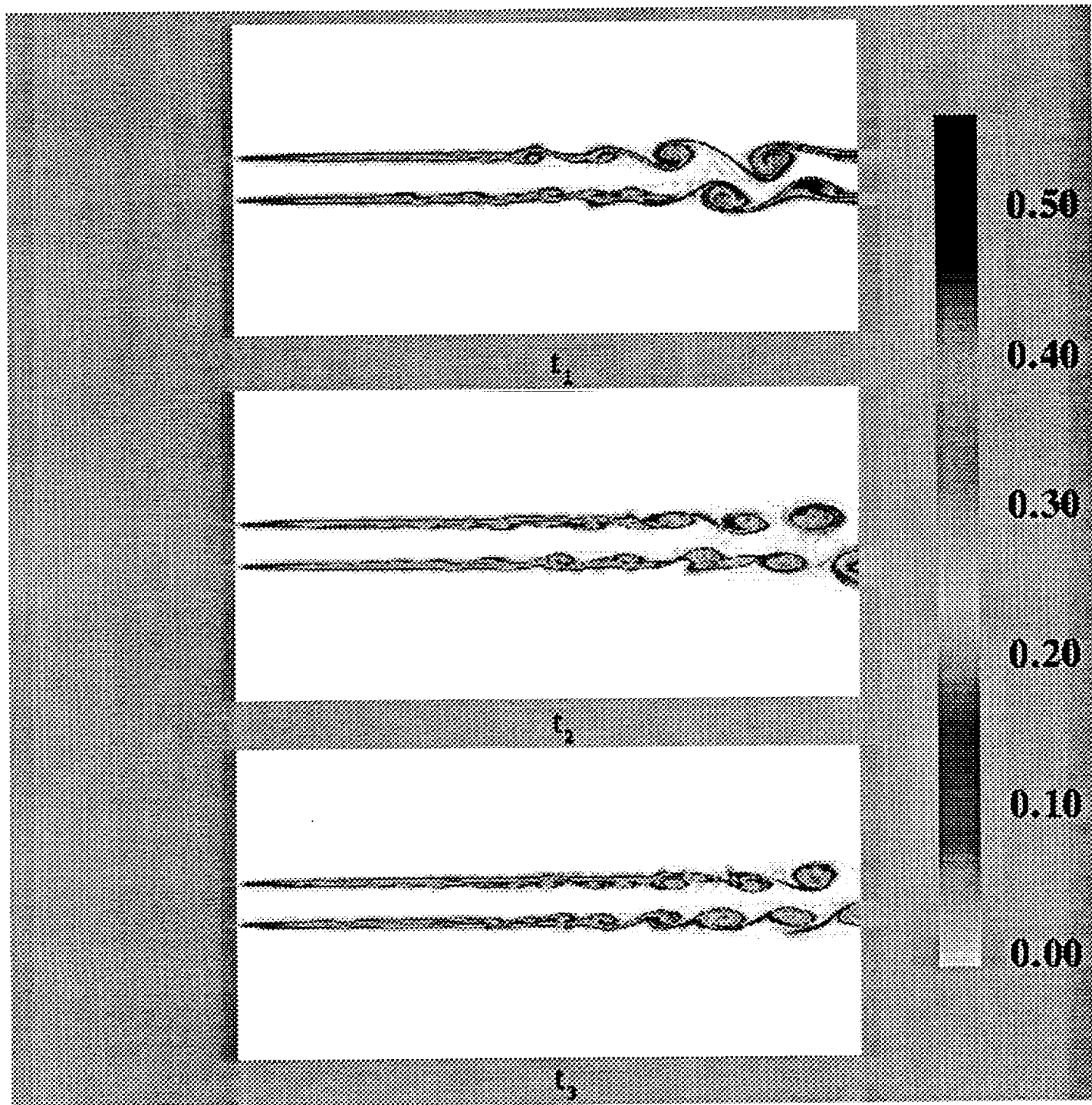
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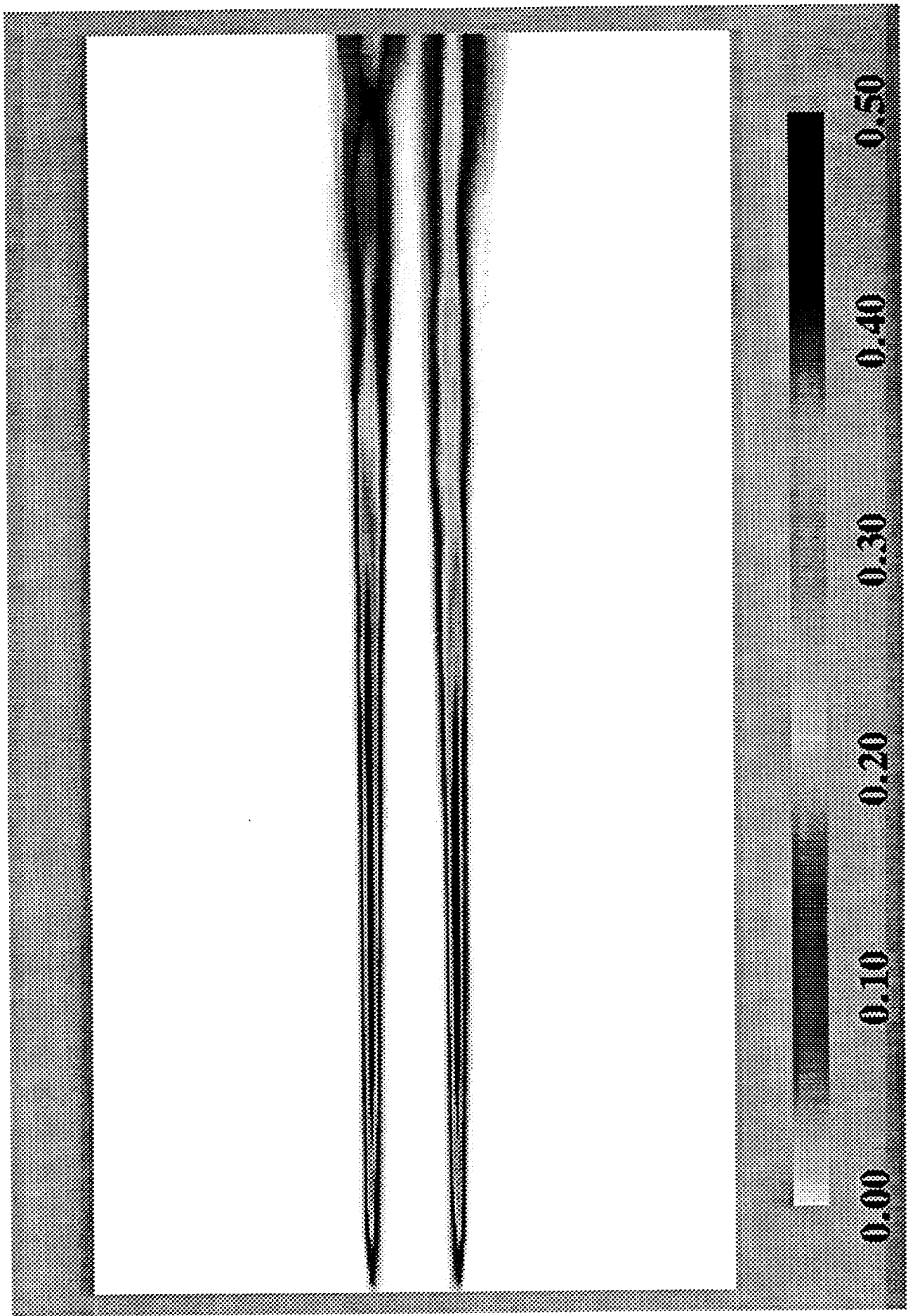
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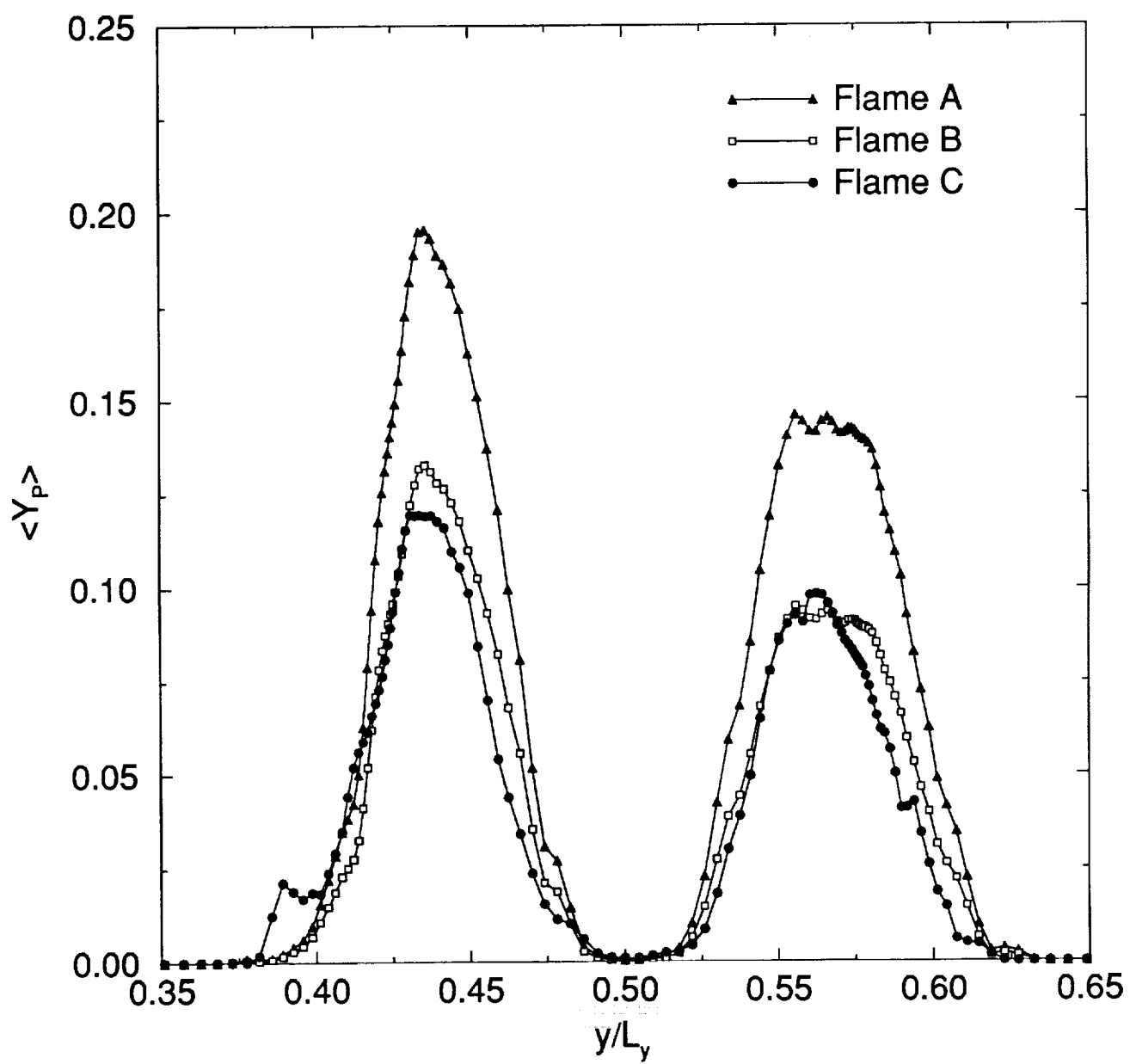
Figure 3
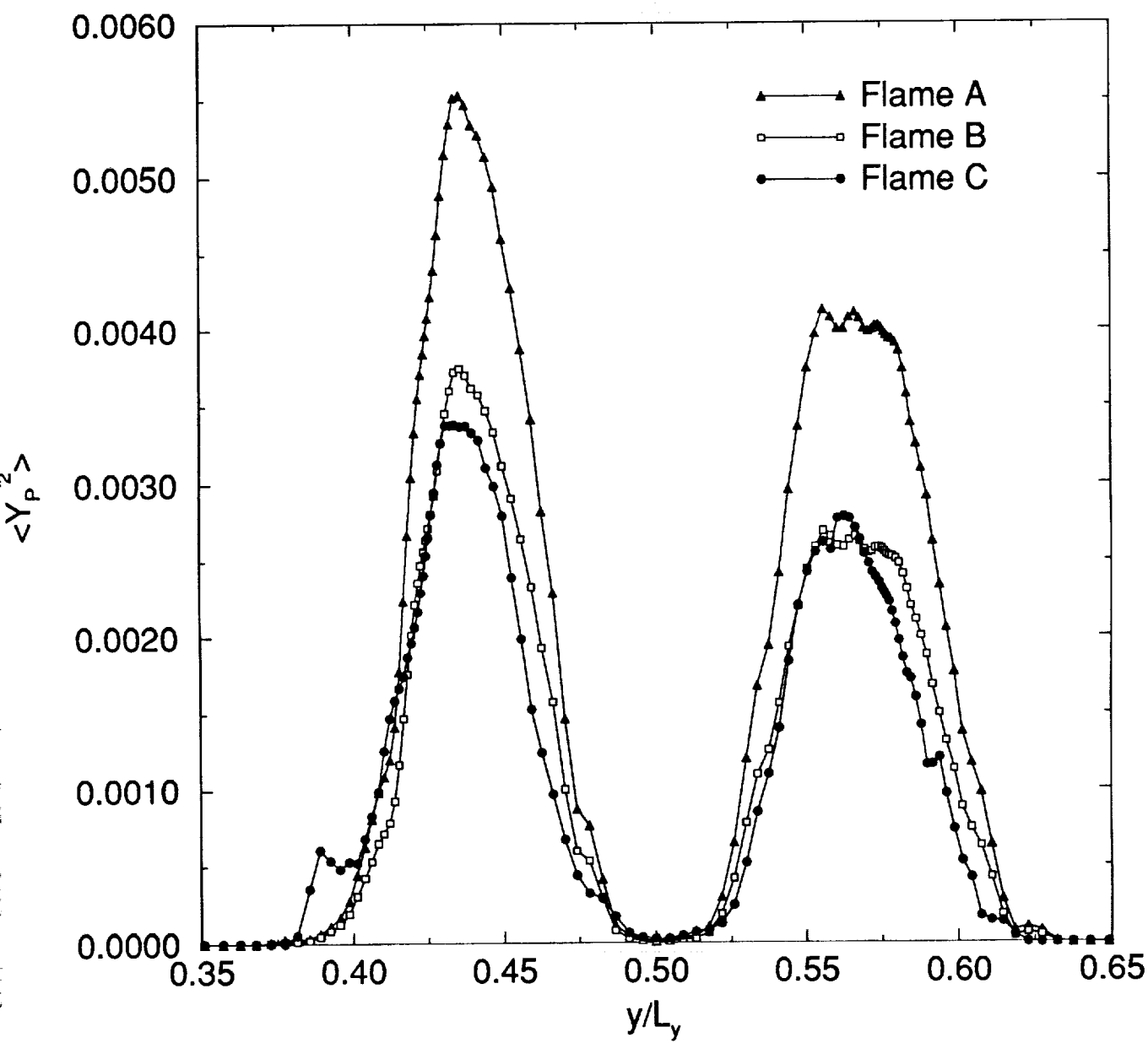
Figure 4
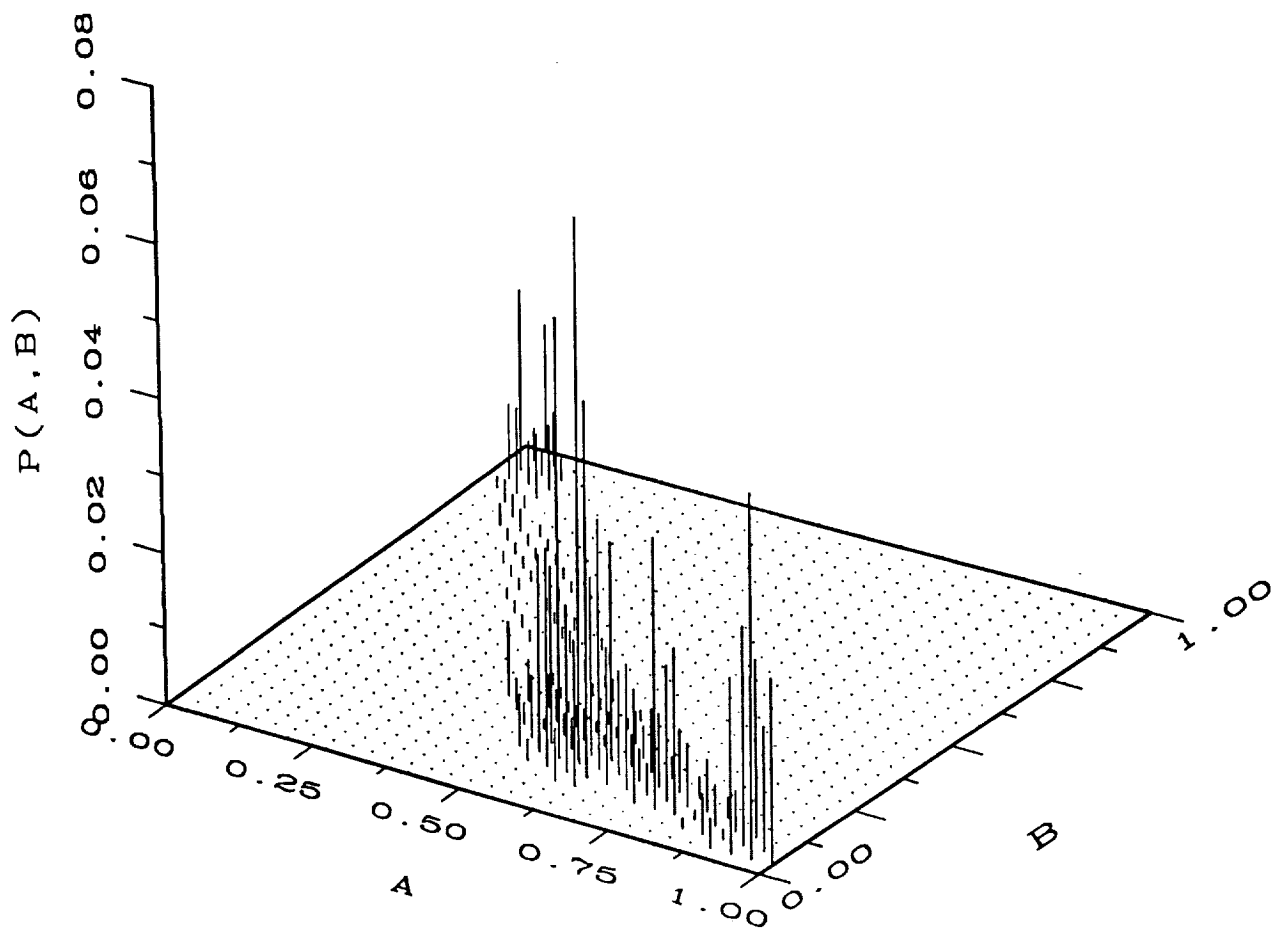
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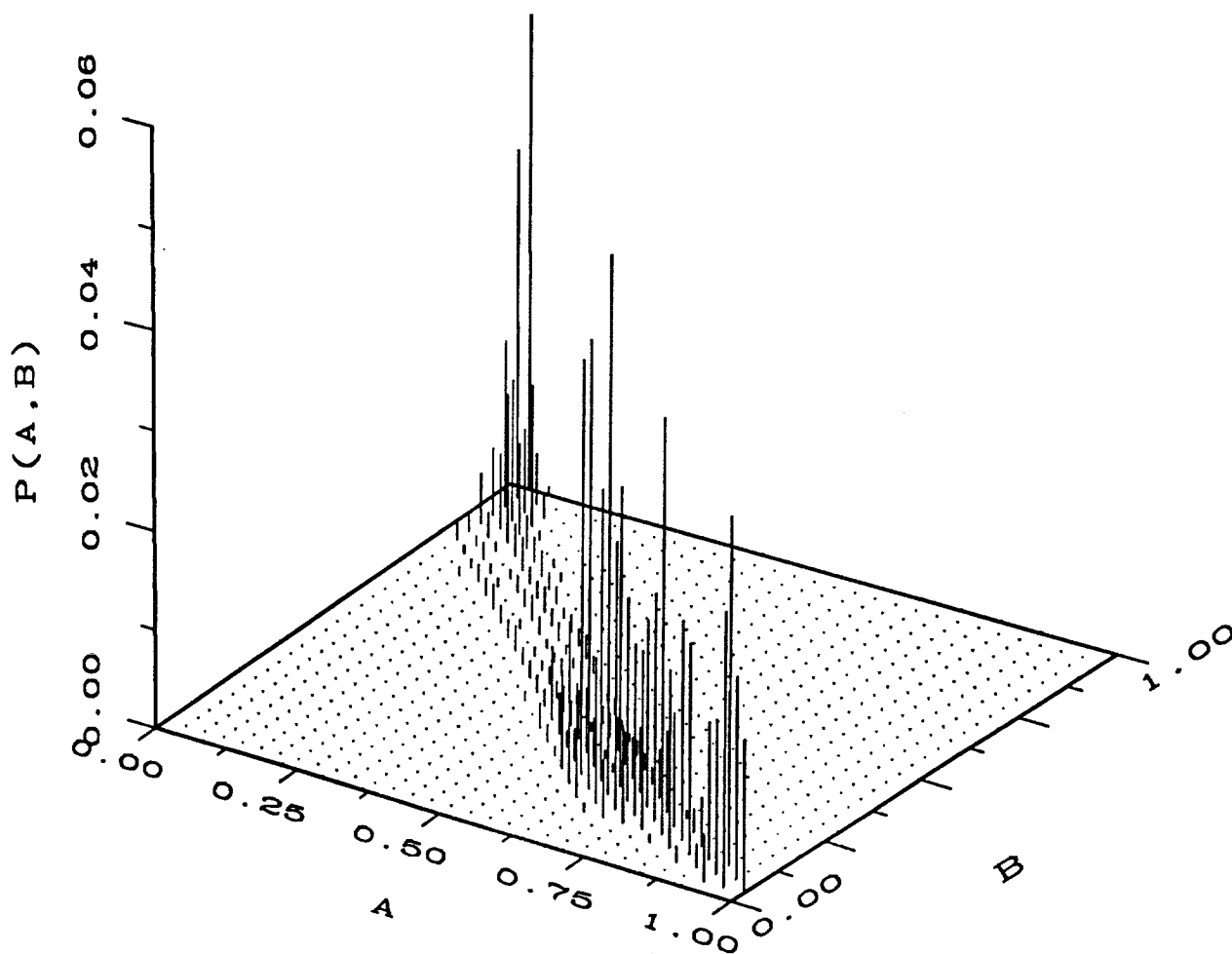
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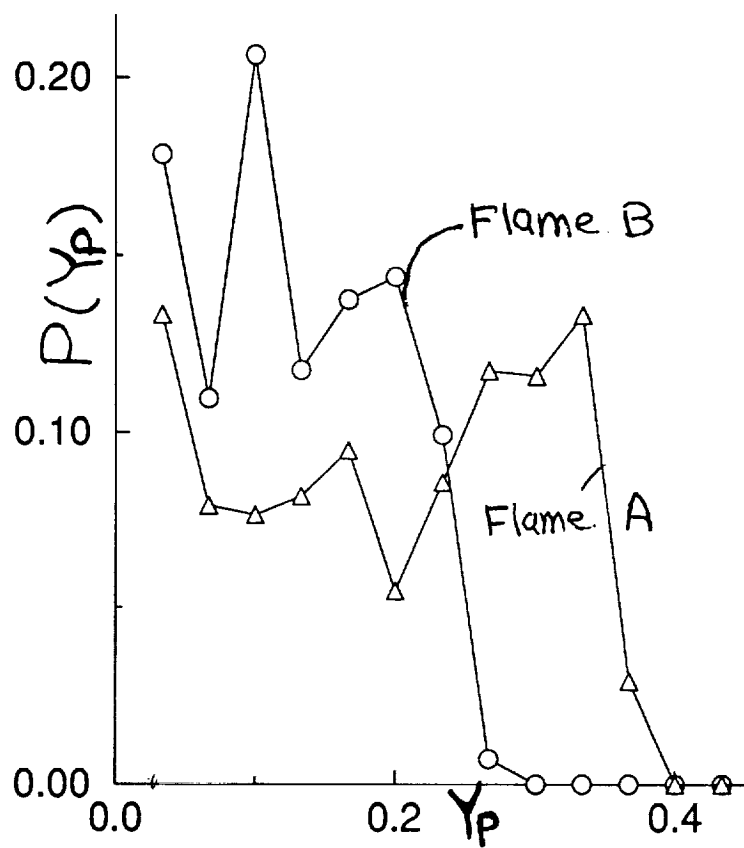
Figure 6

Figure 7a

Da = 0.50, i=125
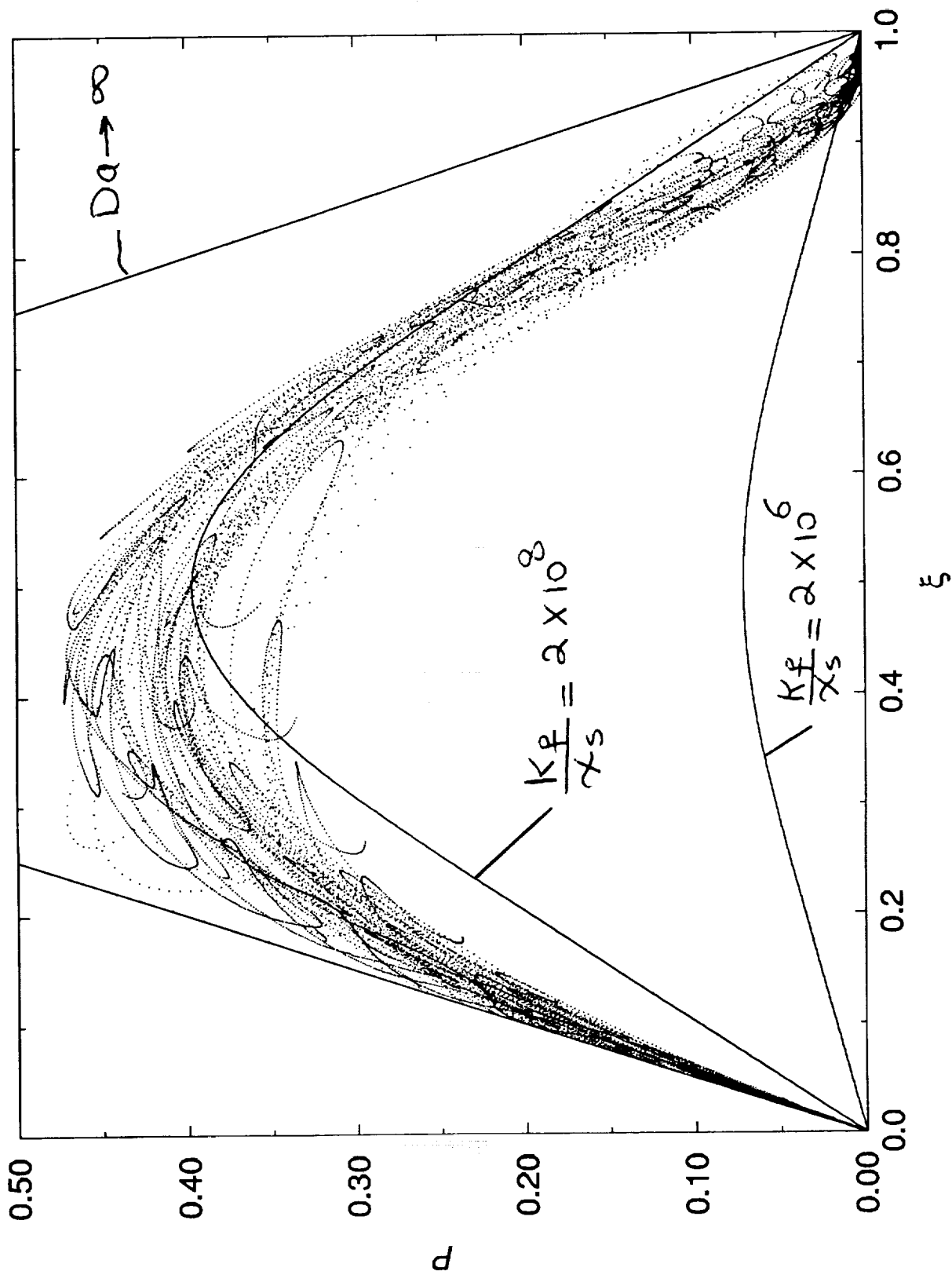
Figure 7(a)

Figure 7b
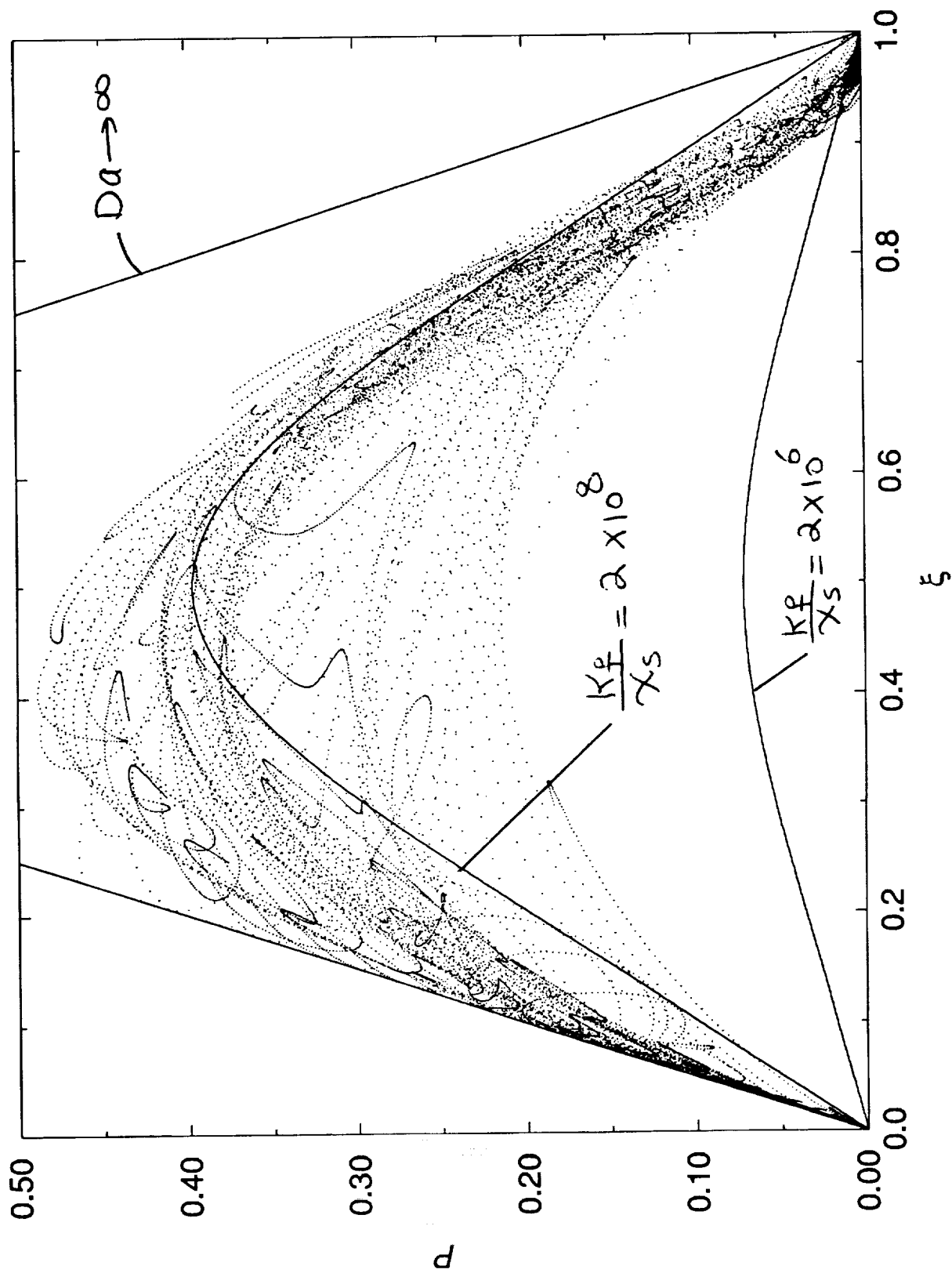
Da = 0.50, i=145

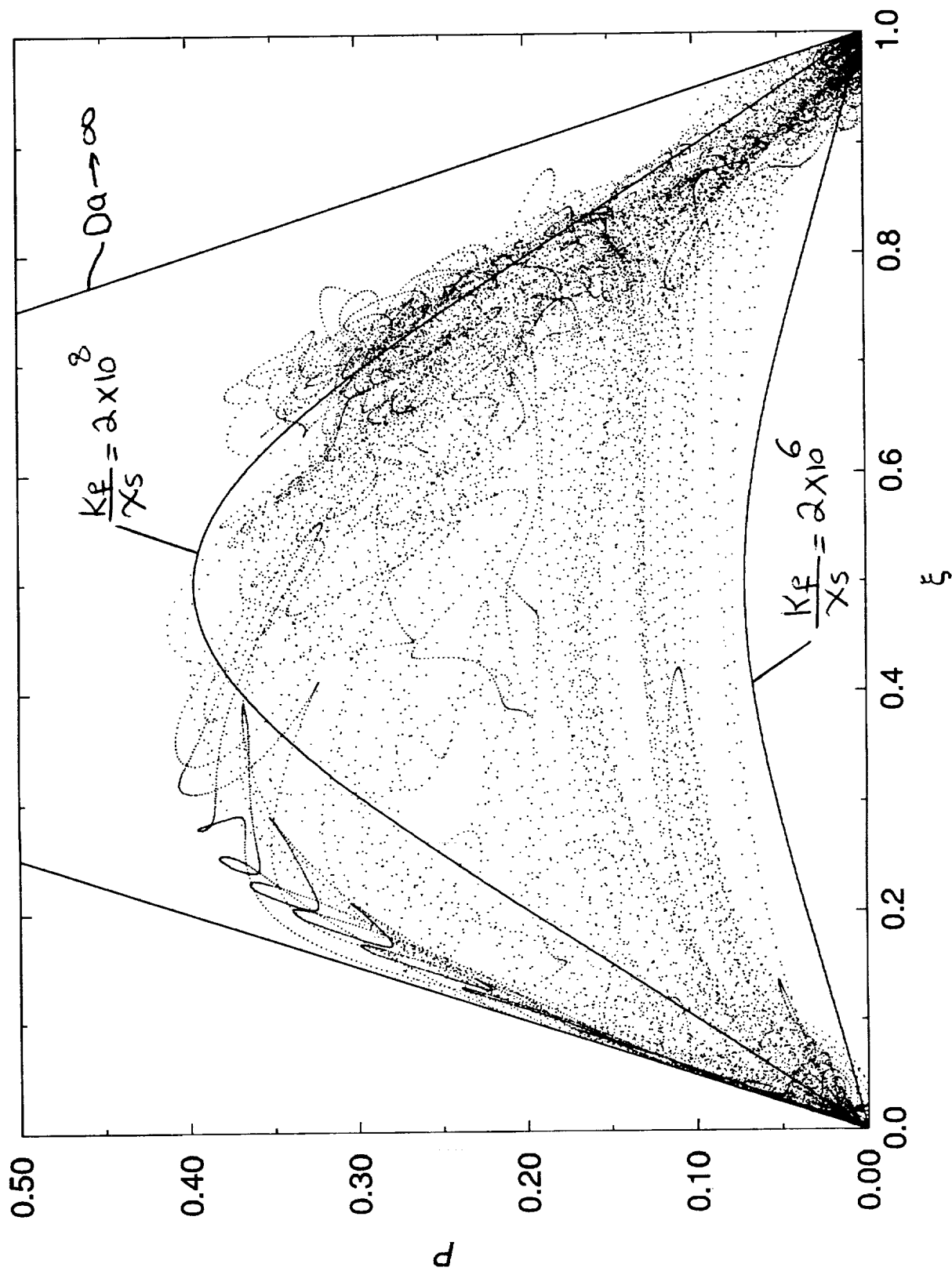Figure 7(b)

Figure 7c

Da = 0.50, i=185

Figure 7(c)

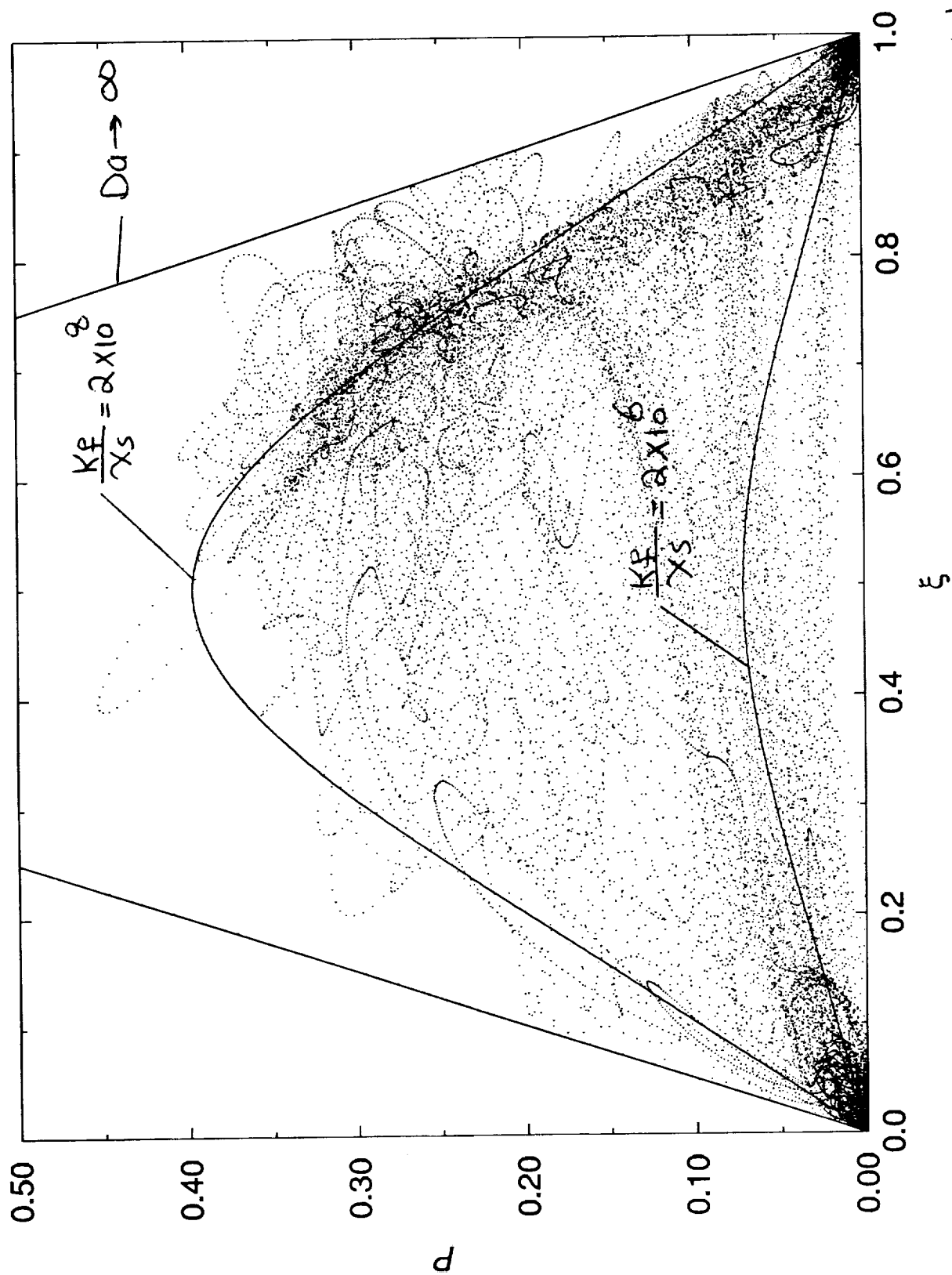# Figure 7d

Da = 0.50, i = 210

Figure 7 (d)

Figure 8a

Da = 0.25, i = 125

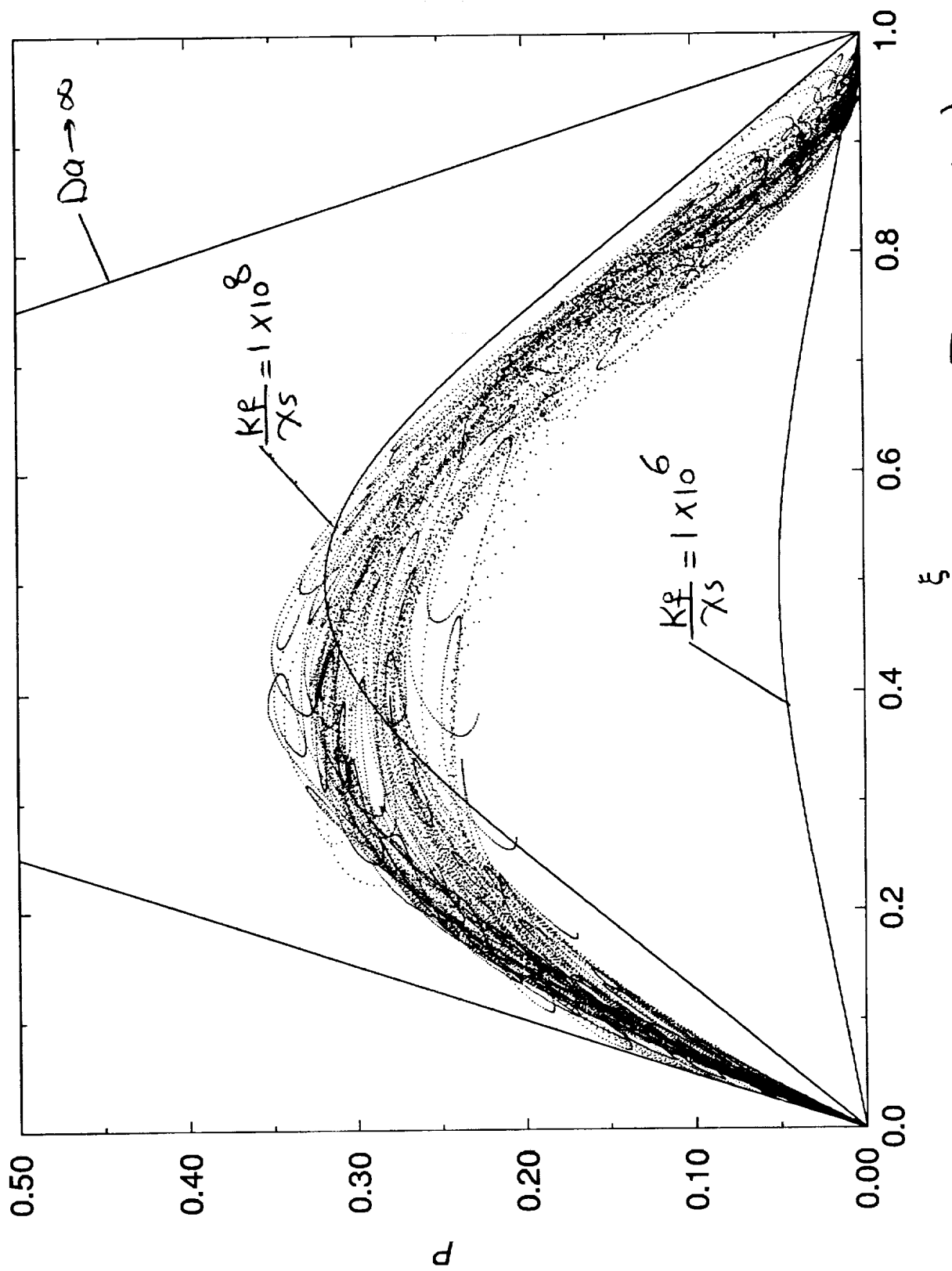Figure 8(a)

Figure 8b

Da = 0.25, i = 145

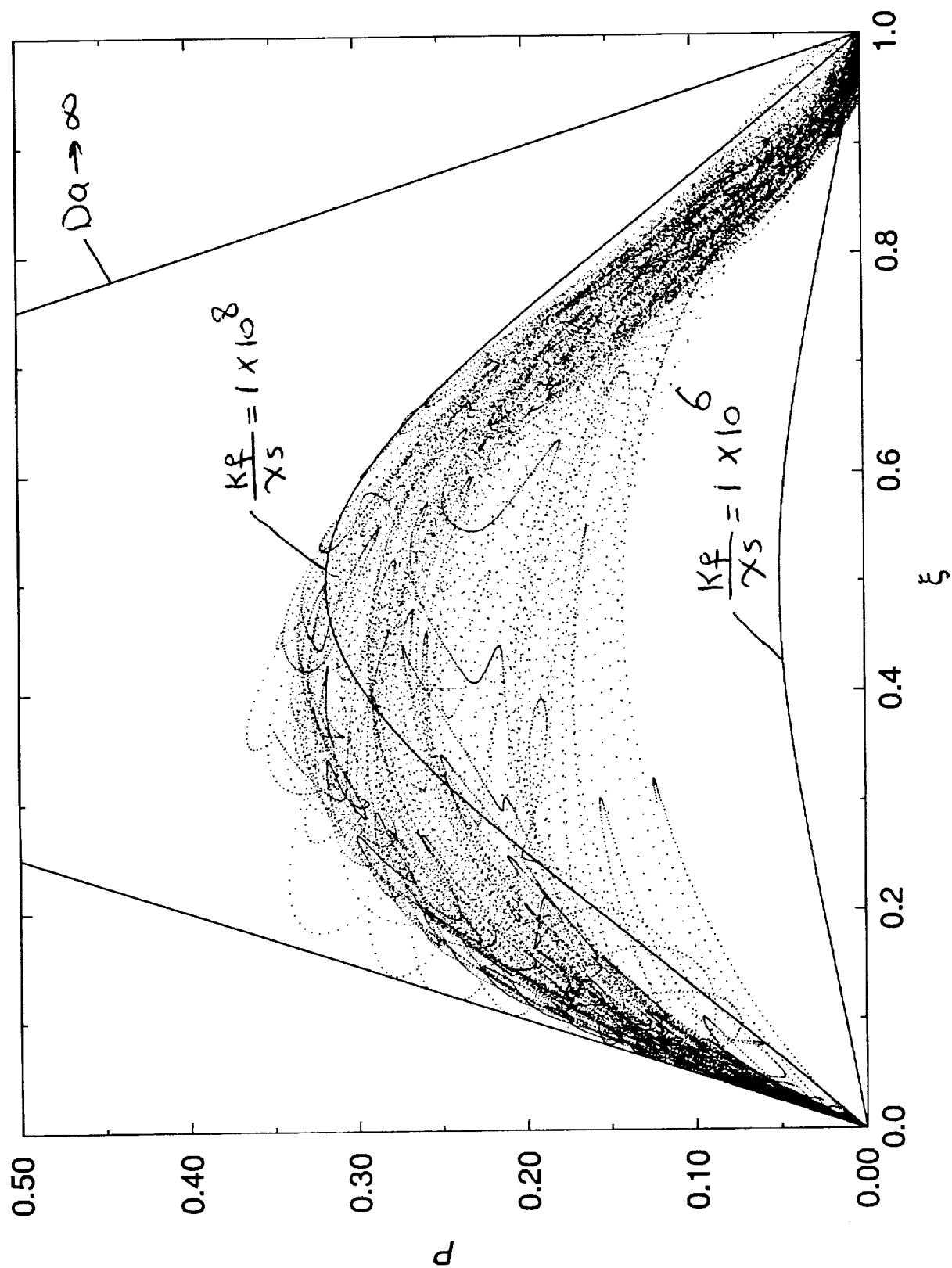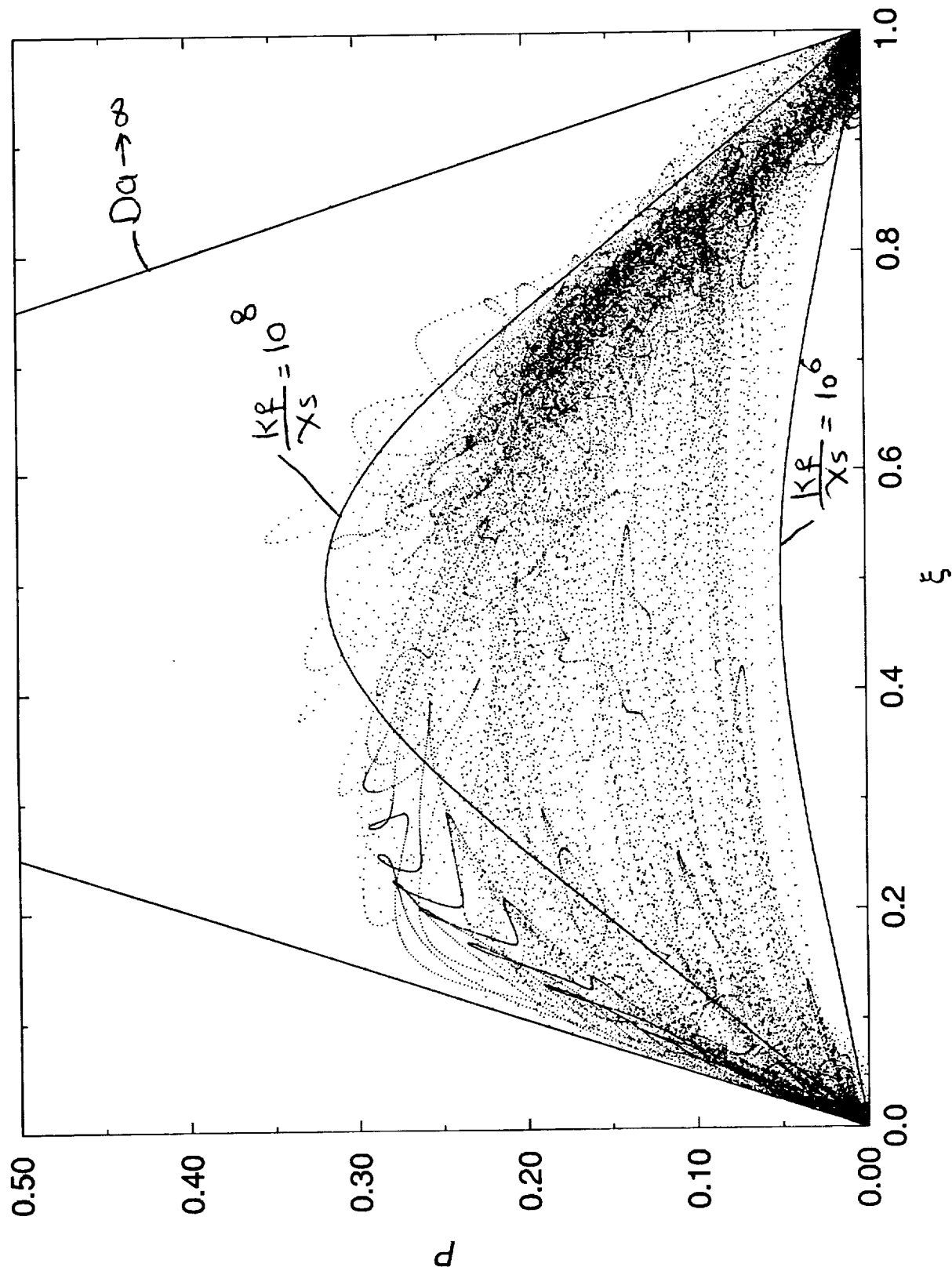Figure 8(b)
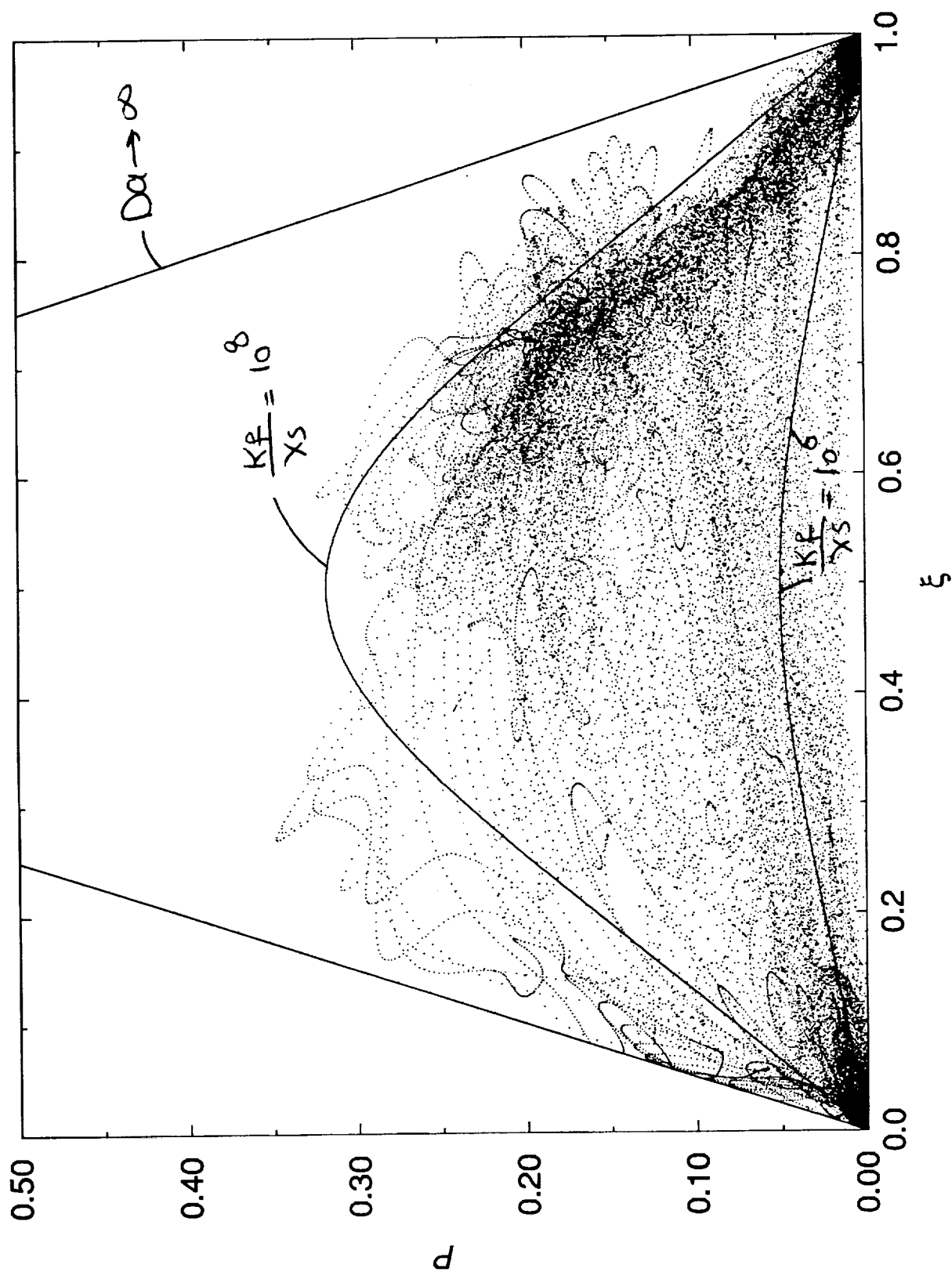
Figure 8c

Da = 0.__, i = 185

Figure 8(c)

Figure 8d

$Da = 0.25, i = 210$

Figure 8(d)

Figure 9

Figure 9